

# ISOTOPIC TRACERS AND NUCLEAR RADIATIONS

With Applications to Biology  
and Medicine





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and Medicine

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WITH APPLICATIONS TO BIOLOGY AND MEDICINE

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## PREFACE

The recent very rapid developments in nuclear physics and the parallel progress in tracer techniques have made it imperative to compile from the vast accumulation of data and experimental procedures some of the material that appears to be most essential to applications of isotopic tracers and nuclear radiations. For the most part, such information is to be found at the present time only in widely scattered literature. Nuclear physicists, to be sure, have access to this information by virtue of their familiarity with the technical literature in the field, but even for the nuclear physicist it is often necessary to refer to many different sources for much of the explicit data he requires. The situation is very much worse for those who are less familiar with nuclear physics but who plan to employ isotopic tracers and high-energy radiations in their research. For such persons, Seaborg and Perlman's "Table of Isotopes" has been literally the only comprehensive source for some of the essential information.

Until recently the compilation of a volume of this kind would have been somewhat futile, since there was too little data that was firmly established and a compilation, once made, would have had to be completely revised and considerably extended six months later. Now, however, it is felt that at least an effort can be made in this direction with the accumulated information derived from the extensive research during and since the war. While it is impossible to present in a single volume the vast amount of material that biologists and physicists in this field would like to have immediately available, it is hoped that the present book, despite its obvious limitations, may find some justification in attempting to provide some of the information for which there seems a pressing demand.

Based on the experience of our laboratory, it was felt that an urgent need could be satisfied if the present volume were prepared in the form of a ready reference in which fundamental data and descriptions of processes and instruments were available in compact form. On the other hand the book was expanded beyond a simple compilation of data and formulas in order to make it more intelligible to those who are less conversant with the terminology and details of nuclear physics and tracer methods. In general, descriptions of processes and instruments have been made rather elementary and with the intention of noting only the essential principles and those data which are important in practice.

The biological and medical aspects of tracer methods, presented by

Dr. Ellsworth C. Dougherty in Part III, is in the form of a survey of those isotopes which have already been used and those which, because of their convenient physical properties and biological significance, may reasonably be employed in future research. No effort has been made to present an exhaustive and detailed account of the biological and medical research since several excellent reviews have already been published, particularly those of Hevesy, Kamen, and Lawrence and Hamilton.

The need for a book covering the fields of tracer methods and nuclear radiations, sufficiently technical to be of use in the laboratory, was early recognized by Col. A. P. Gagge of the Aeromedical Laboratory at Wright Field, Ohio. It was at his suggestion and with the enthusiastic support of Dr. John H. Lawrence that initial preparation of the present volume was undertaken. The first draft, written for the Army Air Forces, was completed in November, 1947, and printed shortly afterward in a limited edition which appeared as A. F. Technical Report No. 5669. Despite the great numbers of errors and omissions, there seemed sufficient interest in this report to justify a comprehensive revision. The original manuscript, consequently, was completely rewritten and extended in scope for the present edition.

Much of the credit for the existence of the book must be given Colonel Gagge and the Army Air Forces, for whose early interest and support we are fully appreciative. From its beginning through to completion, preparation of the book has had the active interest and indispensable support of Dr. Lawrence, to whom the authors owe a great debt of gratitude.

The chapters submitted by the contributing authors, Drs. Ellsworth C. Dougherty, Cornelius A. Tobias, James S. Robertson, Rayburn W. Dunn, and Patricia P. Weymouth, have greatly extended the scope and usefulness of the book. Without their assistance it is doubtful that, even if completed, the book would have served a useful function. We wish especially to thank Dr. Dougherty for the time and effort required for his preparation of the entire biological section of the book, Part III. In addition to the authors cited above we wish to thank Dr. R. L. Dobson for preparing Sec. 19.2 and Dr. M. C. Fishler who began the initial compilation of the bibliography of isotope literature. To aid in identification, the name of each author has been placed at the head of the chapter he submitted. We wish also to acknowledge our indebtedness to Drs. G. T. Seaborg and I. Perlman for permission to use their table of isotopes, to Dr. R. Serber for permission to include the range-energy data calculated by Walter Aron and B. G. Hoffman of the Radiation Laboratory of the University of California, and to the authors and publishers who readily gave permission to reprint many of the graphs and tables.

A considerable portion of the labor in preparing the manuscript was borne

by Miss Jean Smith and Mrs. Frances Schaefer, who typed the greater part of the manuscript, and by Ellis H. Myers, who assisted in preparing the diagrams and graphs. We are especially grateful to Miss M. J. Brandenburg, who not only typed a large part of the manuscript, but also read the entire manuscript and proof, offering many important suggestions and corrections. Many others have been very helpful in the preparation of the manuscript. Among these we wish especially to express our sincere thanks to Drs. R. R. Newell, B. J. Moyer, A. C. Helmholtz, and H. B. Jones for critically reading parts of the manuscript and making many corrections and suggestions.

WILLIAM E. SIRI

Berkeley, Calif.  
June, 1949



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## FOREWORD

The advent of the cyclotron and the nuclear reacting pile has heralded what may be regarded, when viewed by future historians, as the beginning of an entirely new and spectacularly successful era of scientific development. For the physicist, high-energy accelerators have provided the means for unraveling the next and, as yet, the most difficult phase in our progressive understanding of the actual nature of matter. They have made the nucleus accessible to investigation in much the same way that the optical spectrograph permitted an exhaustive study of the atom. The impact of radioactive materials and the half-dozen readily available nuclear radiations on medical research and the biological sciences has been no less profound than for the physical sciences. The rapidly developing tracer techniques have provided a research tool whose power of analysis has only begun to be fully utilized in biology. Although only a few isotopes and nuclear radiations have as yet found a justifiable place in medical investigation, it is difficult at present to predict the medical applications that doubtless will be developed.

In some respects, one of the important results of the development of artificial radioactivity has been the dissolution of the remaining barriers between the biological and physical sciences. Serious students of physics have now found a real challenge in the problems of biology and medicine. They are joining with the biologist in increasing numbers to form efficient research teams with virtually no limits to the fields of experience and knowledge from which they can draw. This book is an example of the concerted effort of physicists, biologists, and chemists. The data and the physical principles and methods so necessary to the intelligent use of tracers and nuclear radiations are to be found for the most part only in the literature of the physical sciences. In that form it is almost inaccessible to the biologist, and it is often inconveniently scattered for the physicist. With this problem in mind, the present volume was prepared in an effort to present the necessary data and methods in compact form. Having observed with considerable interest the development of the book from its beginnings, I feel certain it will serve an important function in supplementing the combined experience of research groups by providing a long needed technical reference. For those persons yet unfamiliar with the physical and biological literature in this field, it would appear to provide much of the fundamental material with which he must familiarize himself before attempting intelligent and useful investigations with isotopic tracers and nuclear radiations.

JOHN H. LAWRENCE



PART I

Isotopes and Nuclear Radiations



## CHAPTER 1

### PROPERTIES OF NUCLEI

**1.1. Stable Isotopes.** Examination of the charge, mass, and abundance of existing isotopes has led to a set of empirical rules governing the structure of stable nuclei so far as the allowed numbers of protons and neutrons in a stable nucleus are concerned [1,2].<sup>1</sup> They are understood in a qualitative way from elementary considerations of the binding energies for various combinations of neutrons and protons and are strongly supported by the behavior of the radioactive isotopes.

The fundamental requirement for stability of nuclei is satisfied when the binding energy is a maximum or, alternatively, the exact mass is a minimum for a given total number of nuclear particles. This may be regarded as equivalent to filling the lowest proton and neutron quantum levels in the nucleus. If the transformation of a proton to a neutron results in a greater binding energy or smaller exact atomic weight, the nucleus is unstable and the transformation will occur through K capture or positron emission. Conversely, positron decay will reduce an excess number of neutrons to protons until the most stable configuration is reached, *i.e.*, until lower lying proton levels are filled. On this basis the isotope rules can be explained and are borne out by the modes of decay of the radioactive isotopes.

Formulation of the isotope rules is simplified with the aid of several designations expressing the relation between numbers of neutrons and protons. Nuclei with the same number of protons  $Z$  but different atomic weights, or  $A - Z$ , are referred to as isotopes; nuclei with the same number of neutrons,  $A - Z$ , but different  $Z$  are isotones; and nuclei with the same  $A$  but different  $Z$  are isobars. Nuclei with even numbers of both protons and neutrons will be designated by (E, E) and those with even protons and odd neutrons by (E, O). Similar definitions hold for the designations (O, E) and (O, O).

1. *In a first approximation, nuclei contain equal numbers of protons and neutrons, i.e.,  $Z = A/2$ .* For the light elements this rule is followed exactly for the most abundant isotope. This can be understood if it is assumed that the nuclear force binding a neutron and proton (pn) is somewhat greater than the attraction between like particles (pp) and (nn). If the reverse were true, one would expect nuclei to consist wholly of neutrons, or neglecting electrostatic repulsion, wholly of protons. Further, if forces between like particles

<sup>1</sup> The numbers in brackets refer to the references at the end of the chapter. The references for this chapter appear on page 14.

(pp) and (nn) exist, they must be nearly equal in magnitude. Assuming that (nn) were greater than (pp), the maximum binding energy and, hence, greatest stability would occur for nuclei with a greater proportion of neutrons. Actually a variation in  $Z = A/2$  of approximately 10 per cent does occur among the isotopes of light elements, but this appears to be insufficient to conclude that a significant difference exists between (nn) and (pp).

The most stable configurations with respect to the purely nuclear forces, therefore, are equal numbers of protons and neutrons. In apparent variance with this, however, is the increase in the proportion of neutrons with increasing atomic weight where, in the heaviest stable isotopes, the ratio of the neutrons to protons reaches a value of 1.6. In these nuclei, however, the repulsive, long-range electrostatic field of the proton becomes a significant factor. The nuclear forces (np), (nn), and (pp) exhibit the property of saturation due to their short range and the finite size of the nucleons. A single particle, therefore, is unaffected by the intrinsically nuclear forces of more distant members of the nucleus, and the total binding energy is then proportional to the number of particles,  $A$ . The electrostatic field, on the other hand, does not show the property of saturation, and consequently each proton is affected by the presence of all other protons in the nucleus. Taken together the protons contribute a total electrostatic energy proportional to  $Z(Z-1)/R$  or  $Z(Z-1)A^{-1/3}$  which tends to diminish the effective total binding energy of the nucleus. In the lightest nuclei the electrostatic energy is less than 0.3 mev per particle, whereas the nuclear forces amount to approximately 8.5 mev per particle. For greater atomic weights the electrostatic energy increases rapidly, and the most stable configuration for a given number of particles is one with a greater proportion of neutrons. A balance between the numbers of neutrons and protons for a given atomic weight is achieved which provides the maximum total binding energy.

2. (E, E) *nuclei are the most stable.* This fact is apparent from both the number and the relative abundance of such nuclei. Of the 278 known stable isotopes, this type includes 164, and where several isotopes of an element of even  $A$  exist the most abundant are (E, E). This is reasonable on the basis of Pauli's exclusion principle. Two particles can occupy the same state, *i.e.*, with identical spatial coordinates, if their spins are different. Pairs of particles in nearly the same quantum state, it can be assumed, form closed shells in which they are strongly bound. If an odd particle is added to the nucleus, it forms an unclosed shell and is weakly bound by interaction with the closed shells.

3. (E, O) and (O, E) *nuclei are about equally stable but less so than (E, E).* Stable nuclei of these types are found to occur in about equal numbers; 58 are (E, O) and 51 are (O, E). A single proton or neutron added to an (E, E) nucleus is less strongly bound through interaction with completed shells at



lower quantum levels as indicated under Rule 2. Nevertheless, such nuclei are stable against K capture or beta emission when the odd particle is added to the next lowest level. The type of stable nucleus that can be formed by the addition of a particle to an (E, E) nucleus of given atomic weight then depends upon the next lowest level to be filled. If this is a proton level but a neutron is added instead, the nucleus is unstable against beta decay and the neutron is transformed to a proton.

4. (O, O) nuclei, with the exception of  $H^2$ ,  $Li^6$ ,  $B^{10}$ , and  $N^{14}$ , are unstable. The existence of only four such stable isotopes indicates immediately the relative instability of this type of nucleus in all but the lightest elements. If each proton-neutron pair occupies the same quantum state, the resulting nucleus is stable and the numbers of protons and neutrons are equal. If, however, the number of neutrons is greater by two or more, they will lie in successively higher levels above the last filled proton level and will transform by beta emission to the lower lying proton level to form nuclei of the type (O, O). Thus, nuclei of this type are stable only if the numbers of protons and neutrons are equal. This condition can be found, however, only for the lightest elements where the electrostatic forces are still small. In heavier elements, stable nuclei containing equal numbers of protons and neutrons cannot exist since the electrostatic repulsion then diminishes the nuclear binding energy as compared with nuclei with the same total number of particles but with a greater proportion of neutrons.

5. For any even  $Z$ , there exists only one or at the most two stable isotopes of odd  $A$ ; if two, they differ by two mass units. Many isotopes of even  $A$  may exist.

6. For any odd  $Z$ , there exists only one or, at most, two isotopes; if two, they differ by two mass units.

7. For any even  $A$ , only two stable isobars may exist and they differ in charge by two units and are even in  $Z$ .

8. For any odd  $A$ , only one stable nucleus exists (no isobars) and its  $Z$  may be even or odd.

A qualitative proof of rules 5, 6, 7, and 8 follows from energy considerations for the various possible combinations of protons and neutrons. A nucleus of mass  $A$  is stable only for that combination of protons and neutrons which provides the maximum binding energy or, alternatively, the minimum exact mass. This can be estimated from the principal terms in the binding-energy formula

$$E = aA - b \frac{(A - 2Z)^2}{A} - \frac{Z^2}{A^{1/2}}$$

$E$  is a maximum when the last two terms are equal in magnitude and opposite in sign. This requires that  $(A - 2Z)/Z \sim A^{3/4}$  or that the excess of neutrons over protons increase as  $A^{3/4}$  for stable nuclei. Further, the binding energy of

nuclei of the same  $A$  varies as  $Z^2$  and is represented therefore by a parabola about the most stable value of  $Z$  located at or near the apex.

For odd  $A$ ,  $Z$  may be even or odd with equal probability as shown by the nearly equal numbers of these stable nuclei. The only isobar that is stable

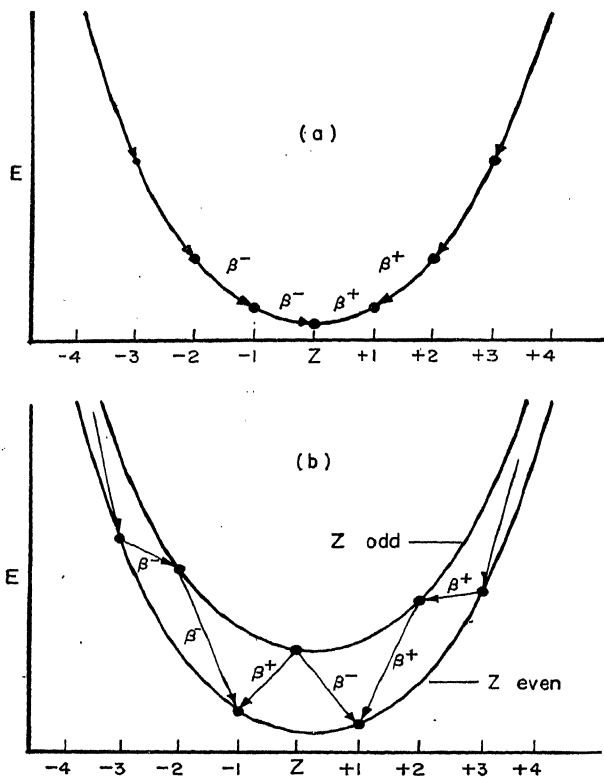


FIG. 1. Schematic diagram representing the cross section of the nuclear energy surface at odd and even isobars. (a) Cross section at odd nuclei. Only the nucleus at  $Z$  is stable, all others move toward this value of  $Z$  (and  $N$ ) by  $\beta^-$ ,  $\beta^+$ , and  $K$  transformation. (b) Cross section at even nuclei. Nuclei at  $Z + 1$  and  $Z - 1$  are stable since transition from one to the other is energetically impossible. All other nuclei are unstable, as indicated by arrows.

is the one lying nearest the apex of the parabola. All other isobars are unstable and move toward the apex either by  $K$  capture or positron or negatron emission until the stable  $Z$  is reached, as shown in Fig. 1. Stated alternatively, isobars with neutrons in energy states higher than the next

unoccupied proton level will transform by beta emission until the lowest levels are filled. The converse holds for isobars with an excess of protons compared with the stable configuration. Such nuclei capture electrons or undergo positron emission transforming protons to neutrons in order to fill lower lying neutron levels.

The binding energy of nuclei with even  $A$  must be represented by two parabolas on the same  $E$  axis: one containing  $(O, O)$  nuclei and a lower curve containing  $(E, E)$  nuclei, as shown in Fig. 1. From the possible combination of  $A$  particles,  $(Z, N)$ ,  $(Z - 1, N + 1)$ ,  $(Z - 2, N + 2)$  etc., and  $(Z + 1, N - 1)$ ,  $(Z + 2, N - 2)$  etc., which can form isobars, those with odd  $Z$  lie on the upper curve and can always transform to a nucleus on the lower curve by beta emission or K capture. By the same processes, isobars lying far up on the  $Z$ -even curve can cross to a lower level on the  $Z$ -odd curve. By successive transformations of this kind the isobar is brought finally to one of two possible stable nuclei occupying the lowest levels on the  $Z$ -even curve. If two points are stable, they must differ by two charge units. These nuclei cannot transform into each other despite a possible energy difference since either beta emission or K capture would take them first to a higher level on the  $Z$ -odd curve, which is energetically impossible, and no process is known for the simultaneous transformation of two charge units without also a change in mass.

An exception to this is found only in the light nuclei  $H^2$ ,  $Li^6$ ,  $B^{10}$ , and  $N^{14}$ . Here, the  $Z$ -even and  $Z$ -odd curves are nearly superimposed because the contribution of the electrostatic field to the binding energy is negligible. The lowest state, therefore, lies at the apex of the  $Z$ -odd curve and  $Z = A - Z = N$ .

**1.2. Mass Defect.** Mass spectrographic measurements of the exact weight of nuclei indicate a consistent variation from the integral values of atomic weight. The difference between the integral atomic weight and the exact mass of an isotope,  $M$ , relative to  $O^{16}$  ( $A = 16.00000$ ) is referred to as the mass defect  $\Delta M$ .

$$\Delta M = M - A$$

The mass defect is positive only for elements lighter than  $O^{16}$  and for the very heaviest elements. In all other cases it is negative. The variation in the mass defect over the entire mass range is of the order of 0.5 per cent or less.

**1.3. Packing Fraction.** The packing fraction  $f$  or mass defect per elementary particle is a quantity most frequently employed in experimental practice to express the deviation of the exact isotope weight from the integral atomic mass number. It is defined by the relation

$$f = \frac{M - A}{A} = \frac{\Delta M}{A}$$

Values of the known packing fractions are given in Table 2 and plotted in Fig. 2. The smooth curve is given by a semiempirical formula calculated by Fowler [3].

$$f = -79.0 + 4.0 \frac{I}{A} + 242.1 \frac{I^2}{A^2} + \frac{179.8}{A^{3/4}} + 7.42 \frac{Z(Z-1)}{A^{3/4}} \times 10^{-4}$$

where  $I = A - 2Z$

**1.4. Binding Energy.** The exact atomic mass of an atom is in all cases less than the mass of an equivalent number of free neutrons and protons plus  $Z$  electrons. This mass difference is given by

$$\Delta M = M_{(A,Z)} - 1.008132Z - 1.00893(A - Z)$$

where 1.008132 is the mass of a proton plus one electron and 1.00893 is the neutron mass. The apparent decrease in the mass of elementary particles bound in a nucleus is exactly equivalent to the total binding energy of the particles. From Einstein's law of equivalence of mass and energy, the binding energy is then  $E = \Delta M c^2$ . One mass unit (mass of  $O^{16}/16$ ) energy equivalent is

$$1 \text{ MU} = 931.05 \text{ mev} = 1.49 \times 10^{-3} \text{ erg}$$

from which the binding energy of a nucleus may be calculated when its exact mass is known.

A semiempirical formula for calculating the binding energy or the mass defect has been derived which is based mainly on the liquid-drop model of the nucleus [1,6,7]. The total binding energy of a nucleus of atomic weight  $A$  and charge  $Z$  may be expressed as the sum of a volume energy, a surface energy, a symmetry (or isotopic spin) energy [7], a coulomb or electrostatic energy, and a somewhat uncertain term involving the fluctuations associated with the even-odd combination of nuclear particles. Evaluation of explicit expressions for the terms has not yet been possible, but the factor to which each term is proportional is readily found as indicated below.

With but few exceptions (deuterium and lithium notably) the average binding energy per nucleon in light and medium nuclei is approximately 8.5 mev. In the heaviest nuclei ( $Z > 82$ ) the binding energy decreases to approximately 6 mev per particle. Over the greater part of the mass range of nuclei the total binding energy is roughly proportional to the number of nucleons or to the nuclear volume. Each particle is influenced by the short-range attractive fields of only those particles next to it and will remain unaffected by more distant particles in the nucleus. To a first approximation then, saturation of the intrinsically nuclear forces leads to a volume energy that is proportional to the number of particles,  $A$ .

Particles lying at the surface of the nucleus would not be expected to

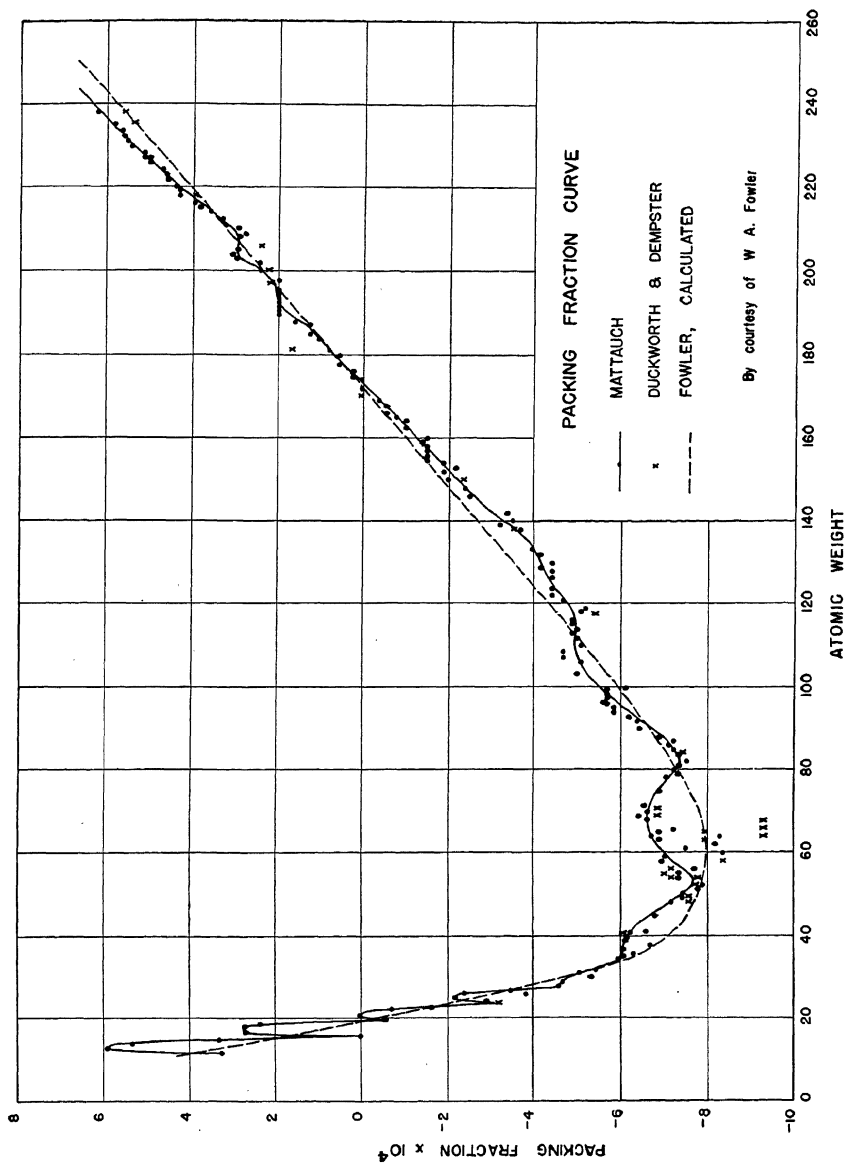


exhibit complete saturation of their nuclear fields as do those lying deeper. The particle is bound to the nucleus by only a portion of its available energy, and the remainder cannot contribute to the total binding energy but gives rise, instead, to a surface tension analogous to that of a liquid drop. The total amount of the energy associated with the surface tension or unsaturated fields must be proportional to the nuclear surface and hence to  $A^{2/3}$  if it is assumed that nuclear matter is incompressible. Referred to as surface energy, this term is deducted from the binding energy first estimated from the nuclear volume. Since the volume-to-surface ratio increases with the radius as  $A^{1/3}$ , the relative magnitude of the surface effect in heavy nuclei is smaller than in light nuclei where it causes an appreciable reduction in the binding energy.

The long-range electrostatic forces due to a uniform volume distribution of  $Z$  protons are taken into account by the third term. These fields cannot be saturated, and since their range is very much greater than nuclear dimensions, each proton is influenced by the repulsive electrostatic fields of the remaining  $Z - 1$  protons. From electrostatics, this term is found to be proportional to  $Z(Z - 1)/R \sim Z(Z - 1)/A^{1/3}$ . The term is relatively unimportant in light nuclei where it amounts to but a small fraction of the energy but increases rapidly with atomic number and becomes, in the heaviest nuclei, a dominant factor in reducing the binding energy and, particularly, the stability against fission (see Fission, Chap. 6).

The symmetry or isotopic-spin [7] energy term is based on the observation that the most stable nuclei are those for which  $Z = A/2$ . From the statistical model of the nucleus [1,2,7] this effect is found to be proportional to  $I^2/A$ , where  $I = A - 2Z = N - Z$  is the isotopic number.

The complete expression for the total binding energy may now be written

$$E = aA + bA^{2/3} + cZ(Z - 1)A^{-1/3} + dI^2A^{-1} + \Delta E$$

where

$$\Delta E = \begin{cases} 0, & \text{for } A \text{ odd} \\ \pm eA^{-2/3}, & \text{for } A \text{ even} \begin{cases} Z \text{ even} \\ Z \text{ odd} \end{cases} \end{cases}$$

The coefficients  $a$ , volume energy,  $b$ , surface energy,  $c$ , electrostatic energy, and  $d$ , symmetry energy, have been determined by fitting the equation to the packing-fraction curve of isotopes for which the masses have been measured with accuracy (see Table 2 and Fig. 2). Table 1 gives three sets of values based on calculations of coefficients in similar formulas for the packing fraction and exact atomic mass. The form of  $\Delta E$  used here is based on that suggested by Deutsch [4]. For a detailed discussion of  $\Delta E$  see reference 7.

The semiempirical binding-energy formula above represents an energy surface in the form of a trough. Stable nuclei lie at points along the bottom

of the trough, while radioactive nuclei are found at points on the sides and move toward the bottom by emission of radiations.

The packing fraction of an atom of atomic weight  $A$  and charge  $Z$  is found from the energy formula by dividing through by  $931A$ . Similarly an estimate of the exact mass of the atom is obtained by dividing the formula by 931 and adding the terms  $1.00893(A - Z) + 1.00812Z$ .

The detailed structure of the observed packing-fraction curve for stable isotopes is not accounted for by the semiempirical formula since it is evaluated

TABLE 1. SEMIEMPIRICAL BINDING ENERGY COEFFICIENTS

Values, mev					Reference
$a$	$b$	$c$	$d$	$e$	
-14.66	15.4	0.602	20.54	....	Mattauch [2]
-15.30	16.75	0.69	22.55	....	Fowler [3]
-13.97	13.0	0.58	38.63	33.5	Deutsch [4]

by an averaging process and no terms of short period are included. A closer fit to the fluctuations in the actual curve must await more detailed information on nuclear structure.

The total binding energy is an important criterion of nuclear stability. At least the lightest nuclei are stable against spontaneous radioactive decay only when their binding energy is greater, *i.e.*, their mass is less, than that for any combination of lighter nuclei containing the same total number of protons and neutrons.

An important application of binding energy is found in the calculation of the exact atomic mass of radioactive nuclei. In the case of beta decay, the energy released (rest mass plus kinetic energy) is exactly equivalent to the difference in the atomic masses of the initial and final atoms, *i.e.*,  $E = c^2(M_Z - M_{Z+1})$ . When a positron is emitted, however, the residual atom is lighter by the equivalent energy carried off by the particle together with its rest energy plus the rest energy of an orbital electron which is also lost.  $E = c^2(M_Z - M_{Z-1} - 2m)$ , where  $m$  is the mass of the positron. Similarly, when an alpha particle is emitted, the energy release is  $E = c^2(M_{A,Z} - M_{(A-4,Z-2)} - 2m)$ . It must be kept in mind, however, that when gamma radiation is emitted its energy must also be taken into account.

**1.5. Nuclear Spin.** The resultant angular momentum or spin of a nucleus is observed in all instances to be either half-integer or integer multiples of  $\hbar/2\pi$ . In particular, the spins of all nuclei with even atomic weight are integral multiples of  $\hbar/2\pi$ , while for nuclei of odd atomic number, the spins are

half-integer value. In most stable, unexcited nuclei the spin is less than  $4h/2\pi$ , and for all nuclei of the even-even type it is zero. The resultant angular momentum of a nucleus is presumably the vector sum of the orbital angular momenta of all the particles and the intrinsic spin of each particle, added according to the vector rules of quantum theory. The orbital angular momentum is always an integer multiple of  $h/2\pi$  if the same quantum conditions hold here as for the orbital electrons. The vector sums therefore are also integers. The spin of both the neutron and proton are known from experimental evidence to be one-half, and if two such particles occupy the same quantum state, their spins must be orientated parallel or antiparallel. The contribution of the particle spins to the total nuclear spin will therefore be an integer or half-integer for even or odd numbers of particles, respectively.

A sufficiently detailed and consistent model of the nucleus has not yet been formulated which will provide the exact magnitude of the spin for a nucleus containing a prescribed number of protons and neutrons. But the qualitative conclusions outlined above regarding the origin and the necessary magnitudes of nuclear spin are borne out by experiment. Further, if the spin of a stable isotope ( $Z, A$ ) is known, it is safe to conclude that the spin of a stable nucleus with which it is isotopic will differ by one-half the number of excess neutrons, *i.e.*,  $i = (i_o/2)(A - A_o)$ , where  $i_o$  is the known spin for the isotope of atomic weight  $A_o$ , and  $A$ , the atomic weight of a stable isotopic nucleus.

The spin of a radioactive nucleus, on the other hand, will depend on its state of excitation and will differ from the ground state, usually by integral units of spin. If a beta particle, for example, is emitted, the nuclear spin always changes by an integral value, including zero, since the ejected beta particle and neutrino each have an intrinsic spin of one-half. In any nuclear reaction, spin and angular momentum must be conserved as well as mass and energy. Known values of nuclear spins in units of  $h/2\pi$  are given in Table 3 page 22 for stable nuclei.

**1.6. Magnetic Dipole Moment.** Assuming the particles within a nucleus to be in motion, it is to be expected that a magnetic field will be produced by the current distribution of at least the charged particles. A single proton moving in a circular orbit with a frequency  $\nu$  and angular momentum  $lh/2\pi$  represents a circulating current of magnitude equal to  $i = e\nu$  and produces a field equivalent to a magnetic shell with a magnetic moment of

$$\mu = \frac{leh}{4\pi Mc} \quad l\mu_o$$

The unit  $\mu_o$  is referred to as the nuclear magneton. Because of the relative magnitudes of the masses of the electron and proton it is smaller than the Bohr magneton by the factor 1,840.



Without a more detailed knowledge of the structure and the motions of particles in nucleus, the magnetic moment must be determined experimentally. Both the proton and neutron themselves have a magnetic moment which also must be determined experimentally because the charge distribution in elementary particles is not known. Although the magnetic moment of the electron is determined directly by its spin and angular momentum, this is not true of the proton and neutron, neither of which have magnetic moments of one nuclear magneton, the value suggested by the ratio of their masses to that of the electron. This discrepancy between nuclear angular moments and the corresponding magnetic moments is taken into account by the formal introduction of a nuclear  $g$  factor which is defined as the ratio of the magnetic moment in units of nuclear magnetons,  $\mu_o$ , to the angular momentum in units of  $\hbar/2\pi$ . Thus, the magnetic moment in units of nuclear magnetons is  $\mu = ig$ , where  $i$  is the nuclear spin in units of  $\hbar/2\pi$ . Most experimental methods give the nuclear  $g$  factor directly, and if the spin is known,  $\mu$  is given the simple product as indicated above.

Known experimental values of  $\mu$  are given in Table 3.

**1.7. Electric Quadrupole Moment.** From the theory of electrostatics it is known that the potential field of an arbitrary charge distribution can be expressed in a series of terms of the form

$$V = \frac{eZ}{r} + \frac{ed}{r^2} P_1(\cos \theta) + \frac{q}{r^3} P_2(\cos \theta) + \dots$$

where  $P_i(\cos \theta)$  are Legendre polynomials. At large distances compared with the dimensions of the charge distribution, only the first term, representing a spherically symmetric or coulomb field, is important since all other terms diminish as  $1/r^2$  or faster. Thus, at distances large compared with the nuclear radius the electrostatic field is equivalent to a point source of charge  $eZ$ . The second term, representing an electric dipole, does not exist for nuclei since it vanishes for radially symmetric charge distributions containing charge of only one sign.

The third term, representing the electric quadrupole, may, however, exist for nuclei in which protons are not distributed throughout the nucleus with strict spherical symmetry. A small contribution to the electric field is then made by the quadrupole moment which in effect alters the spherically symmetric form of the stronger coulomb field but only within a distance of a few times the nuclear radius. It is reasonable to assume in a first approximation that the asymmetry takes the simplest form of distortion of a sphere which is that of an ellipsoid of rotation. Then from electrostatics the nucleus will be cigar-shaped or a prolate spheroid when  $q$  is positive, and platter-shaped or an oblate spheroid when  $q$  is negative.

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4. DEUTSCH, M.: "Science and Engineering of Nuclear Power," Vol. 1, C. Goodman, ed., Addison-Wesley Press Inc., Cambridge, Mass., 1947.
5. HEISENBERG, W.: Rapport du VII Congrès Salvay, Paris, 1934.
6. WEIZSACKER, C. F. V.: *Physik. Z.*, **96**, 431 (1935).
7. FEENBERG, E.: *Rev. Mod. Phys.*, **19**, 239 (1947).

TABLE 2. ATOMIC WEIGHT AND NATURAL ABUNDANCE OF ISOTOPES

An asterisk (\*) indicates that the isotope is radioactive. The relative abundances of most medium and heavy elements are subject to some uncertainty since values reported by various investigators differ by as much as 50 per cent for the rare isotopes and as much as 10 per cent for the more abundant.

Isotope			Abundance, per cent	Atomic weight $O^{16} = 16.000000$ Error $\times 10^6$ mu	Packing fraction $\times 10^4$ mu	Chemical atomic weight
Z	El.	A				
1	H	1	99.9844	1.008123 $\pm$ 0.6	81.2	1.00785
		2	0.0156	2.014708 $\pm$ 1.1	73.6	
2	He	3	$1.3 \times 10^{-4}$	3.01700 $\pm$ 4.	56.7	4.003
		4	$\sim 100$ .	4.00390 $\pm$ 3.	9.8	
3	Li	6	7.39	6.01697 $\pm$ 5.	20.3	6.940
		7	92.61	7.01822 $\pm$ 6.	26.0	
4	Be	9	100.	9.01503 $\pm$ 6.	16.4	9.02
5	B	10	18.83	10.01677 $\pm$ 8.	16.7	10.82
		11	81.17	11.01244 $\pm$ 19.	11.3	
6	C	12	98.9	12.00382 $\pm$ 4.	3.2	12.010
		13	1.1	13.007581 $\pm$ 2.5	5.8	
7	N	14	99.62	14.00751 $\pm$ 4.	5.4	14.008
		15	0.38	15.004934 $\pm$ 3.	3.3	
8	O	16	99.76	16.000000	0.0	16.00000
		17	0.04	17.00450 $\pm$ 6.	2.7	
		18	0.20	18.0049 $\pm$ 40.	2.7	
9	F	19	100.	19.00452 $\pm$ 17.	2.4	19.00
10	Ne	20	90.00	19.99877 $\pm$ 10.	-0.6	20.183
		21	0.27	20.99963 $\pm$ 22.	-0.2	
		22	9.73	21.99844 $\pm$ 36.	-0.7	
11	Na	23	100.	22.99618 $\pm$ 31.	-1.7	22.997
12	Mg	24	78.60	23.9924 $\pm$ 60.	-3.2	24.32
		25	10.11	24.9938 $\pm$ 90.	-2.5	
		26	11.29	25.9898 $\pm$ 50.	-3.9	
13	Al	27	100.	26.9899 $\pm$ 80.	-3.7	26.97
14	Si	28	92.28	27.9866 $\pm$ 60.	-4.6	28.086
		29	4.67	28.9866 $\pm$ 60.	-4.5	
		30	3.05	29.9832 $\pm$ 90.	-5.7	
15	P	31	100.	30.9842 $\pm$ 50.	-5.2	30.98
16	S	32	95.1	31.98089 $\pm$ 7.	-6.0	32.06
		33	0.74	32.9800 $\pm$ 60.	-5.2	
		34	4.2	33.97710 $\pm$ 35.	-6.5	
		36	0.016	35.978 $\pm$ 100.	-6.1	
17	Cl	35	75.4	34.97867 $\pm$ 21.	-6.1	35.457
		37	24.6	36.97750 $\pm$ 14.	-6.1	
18	A	36	0.307	35.9780 $\pm$ 100.	-6.8	39.944
		38	0.061	37.974 $\pm$ 250.	-6.8	
		40	99.632	39.97504 $\pm$ 26.		
19	K	39	93.44	38.9747	-6.5	39.096
		40*	0.012	39.9760 $\pm$ 100.	-6.0	
		41	6.55	40.974	-6.3	
20	Ca	40	96.96	39.9753 $\pm$ 150.	-6.2	40.08
		42	0.64	41.9711	-6.8	

TABLE 2. ATOMIC WEIGHT AND NATURAL ABUNDANCE OF ISOTOPES—(Continued)

Isotope			Abundance, per cent	Atomic weight $O^{16} = 16.000000$ . Error $\times 10^6$ mu	Packing fraction $\times 10^4$ mu	Chemical atomic weight
Z	El.	A				
20	Ca	43	0.15	42.9723	-6.3	
		44	2.06			
		46	0.0033			
		48	0.185			
21	Sc	45	100.	44.9689 $\pm 100.$	-6.8	45.10
22	Ti	46	7.95	45.9661 $\pm 100.$	-7.4	47.90
		47	7.75	46.9647 $\pm 100.$	-7.5	
		48	73.45	47.9651 $\pm 50.$	-7.3	
		49	5.51	48.9646 $\pm 60.$	-7.2	
		50	5.34	49.9646 $\pm 40.$	-7.6	
23	V	51	100.	50.9577 $\pm 50.$	-8.3	50.95
24	Cr	50	4.49	49.96443 $\pm 3.9$	-7.11	52.01
		52	83.75	51.95589 $\pm 4.4$	-8.47	
		53	9.55	52.95527 $\pm 4.4$	-8.44	
		54	2.38	53.95427 $\pm 4.8$	-8.47	
25	Mn	55	100.	54.965	-6.4	54.93
26	Fe	54	5.81	53.95774 $\pm 4.8$	-7.83	55.84
		56	91.66	55.95340 $\pm 2.7$	-8.32	
		57	2.20	56.95485 $\pm 5.2$	-7.92	
		58	0.33	57.95091 $\pm 4.9$	-8.46	
27	Co	59	100.	.....	.....	58.94
28	Ni	58	67.76	57.95939 $\pm 40.$	-7.0	58.69
		60	26.16	59.94951 $\pm 31.$	-8.4	
		61	1.21	60.9537 $\pm 150.$	-7.6	
		62	3.66	61.94928 $\pm 40.$	-8.2	
		64	1.16	63.94712 $\pm 56$	-8.4	
29	Cu	63	69.09	62.956	Mean	63.542
		65	30.91	64.955	-8.13	
30	Zn	64	48.89	63.95365 $\pm 6.6$	-7.24	65.38
		66	27.81	65.94676 $\pm 4.2$	-8.07	
		67	4.07	66.94826 $\pm 3.8$	-7.71	
		68	18.61	67.94885 $\pm 6.5$	-7.53	
		70	0.620	69.9461 $\pm 17.$	-7.69	
31	Ga	69	61.2	68.955	-6.5	69.72
		71	38.8	70.953	-6.6	
32	Ge	70	20.55	.....	.....	72.60
		72	27.37	.....	.....	
		73	7.61	.....	.....	
		74	36.74	.....	.....	
		76	7.67	.....	.....	
33	As	75	100.	.....	.....	74.91
34	Se	74	0.87	.....	.....	78.96
		76	9.02	.....	.....	

TABLE 2. ATOMIC WEIGHT AND NATURAL ABUNDANCE OF ISOTOPES—(Continued)

Isotope			Abundance, per cent	Atomic weight $O^{16} = 16.000000$ . Error $\times 10^5$ mu	Packing fraction $\times 10^4$ mu	Chemical atomic weight
Z	El.	A				
34	Se	77	7.58			
		78	23.52			
		80	49.82			
		82	9.19			
35	Br	79	50.53	$79.94438 \pm 5.0$	-7.04	79.916
		81	49.47	$80.94228 \pm 3.8$	-7.12	
36	Kr	78	0.342	77.945	-7.0	83.7
		80	2.223			
		82	11.50	81.939	-7.5	
		83	11.48			
		84	57.02	83.938	-7.3	
		86	17.43	85.939	-7.1	
37	Rb	85	72.8	.....	.....	85.48
		87*	27.2			
38	Sr	84	0.56	.....	.....	87.63
		86	9.86			
		87	7.02			
		88	82.56			
39	Y	89	100.	.....	.....	88.92
40	Zr	90	51.51	.....	.....	91.22
		91	11.27			
		92	17.14			
		94	17.30			
		96	2.78			
41	Cb	93	100.	.....	.....	92.91
42	Mo	92	15.86	.....	.....	95.95
		94	9.12			
		95	15.70	94.945	-5.8	
		96	16.50	95.946	-5.6	
		97	9.45	96.945	-5.8	
		98	23.75	97.944	-5.7	
		100	9.63			
43	Tc	99				
44	Ru	96	5.68	95.946	-5.7	101.7
		98	2.22			
		99	12.81	98.944	-5.7	
		100	12.70			
		101	16.98			
		102	31.34			
		104	18.27			
45	Rh	103	100.	.....	.....	102.91
46	Pd	102	0.8	.....	.....	106.7
		104	9.3	103.946	-5.2	

TABLE 2. ATOMIC WEIGHT AND NATURAL ABUNDANCE OF ISOTOPES—(Continued)

Isotope			Abundance, per cent	Atomic weight $O^{16} = 16.000000$ . Error $\times 10^5$ mu	Packing fraction $\times 10^4$ mu	Chemical atomic weight
Z	El.	A				
46	Pd	105	22.6	104.945	-5.2	
		106	27.2	105.945	-5.2	
		108	26.8	107.943	-5.2	
		110	13.5	109.942	-5.2	
47	Ag	107	51.35	106.948	-4.8	107.880
		109	48.65	108.947	-4.8	
48	Cd	106	1.215	.....	.....	112.41
		108	0.875			
		110	12.39			
		111	12.75			
		112	24.07			
		113	12.26			
		114	28.86			
		116	7.58			
49	In	113	4.23	.....	.....	114.76
		115	95.77			
50	Sn	112	0.90	.....	.....	118.70
		114	0.61			
		115	0.35			
		116	14.07	115.942	-5.0	
		117	7.54			
		118	23.98	117.939	-5.1	
		119	8.62	118.938	-5.2	
		120	33.03	119.94	-6.	
		122	4.78	121.944	-4.6	
		124	6.11	123.943	-4.6	
51	Sb	121	57.25	.....	.....	121.76
		123	42.75			
52	Te	120	0.091	.....	.....	127.64
		122	2.49			
		123	0.89			
		124	4.63			
		125	7.01			
		126	18.72			
		128	31.72			
		130	34.46			
53	I	127	100.	.....	.....	126.92
54	Xe	124	0.094	.....	.....	131.3
		126	0.088			
		128	1.90			
		129	26.23	128.946	-4.2	
		130	4.07			
		131	21.17			

TABLE 2. ATOMIC WEIGHT AND NATURAL ABUNDANCE OF ISOTOPES—(Continued)

Isotope			Abundance, per cent	Atomic weight $O^{16} = 16.000000$ . Error $\times 10^6$ mu	Packing fraction $\times 10^4$ mu	Chemical atomic weight
Z	El.	A				
54	Xe	132	26.96	131.946	-4.4	
		134	10.54			
		136	8.95			
55	Cs	133	100.	.....	.....	132.91
56	Ba	130	0.101	.....	.....	137.36
		132	0.097			
		134	2.42			
		135	6.59			
		136	7.81			
		137	11.32			
		138	71.66			
57	La	138	0.089	.....	.....	138.92
		139	99.911	138.955	-3.2	
58	Ce	136	0.193	.....	.....	140.13
		138	0.250			
		140	88.48			
		142	11.07			
59	Pr	141	100.	.....	.....	140.92
60	Nd	142	27.27	.....	.....	144.27
		143	12.26			
		144	23.95			
		145	8.27			
		146	17.06	145.960	-2.8	
		148	5.66	147.961	-2.7	
		150	5.53	149.967	-2.2	
61	Pm		.....	.....	.....	150.43
62	Sm	144	3.16			
		147	15.07			
		148*	11.27			
		149	13.85			
		150	7.47			
		152	26.63			
		154	22.53			
63	Eu	151	47.77	.....	.....	152.0
		153	52.23			
64	Gd	152	0.21	.....	.....	156.9
		154	2.14			
		155	14.86	154.977	-1.5	
		156	20.61	155.976	-1.5	
		157	15.66	156.976	-1.5	
		158	24.75			
		160	21.77			
65	Tb	159	100.	.....	.....	159.2

TABLE 2. ATOMIC WEIGHT AND NATURAL ABUNDANCE OF ISOTOPES—(Continued)

Isotope			Abundance, per cent	Atomic weight $O^{16} = 16.000000$ Error $\times 10^6$ mu	Packing fraction $\times 10^4$ mu	Chemical atomic weight
Z	El.	A				
66	Dy	156	0.0524	.....	.....	162.46
		158	0.0902			
		160	2.294			
		161	18.88			
		162	25.53			
		163	24.97			
		164	28.18			
67	Ho	165	100.	.....	.....	164.935
68	Er	162	0.1	.....	.....	167.2
		164	1.5			
		166	32.9			
		167	24.4			
		168	26.9			
		170	14.2			
69	Tm	169	100.	.....	.....	169.4
70	Yb	168	0.06	.....	.....	173.04
		170	4.21			
		171	14.26			
		172	21.49			
		173	17.02			
		174	29.58			
		176	13.38			
71	Lu	175	97.5	.....	.....	174.99
		176*	2.5			
72	Hf	174	0.18	.....	.....	178.6
		176	5.30			
		177	18.47			
		178	27.10			
		179	13.84			
		180	35.11			
73	Ta	181	100.	.....	.....	180.88
74	W	180	0.135	.....	.....	183.88
		182	26.41			
		183	14.40			
		184	30.64			
		186	28.41			
75	Re	185	37.07	.....	.....	186.31
		187	62.92			
76	Os	184	0.018	.....	.....	190.2
		186	1.59			
		187	1.64			
		188	13.3			
		189	16.1			



TABLE 2. ATOMIC WEIGHT AND NATURAL ABUNDANCE OF ISOTOPES—(Continued)

Isotope			Abundance, per cent	Atomic weight $O^{16} = 16.000000$ . Error $\times 10^6$ mu	Packing fraction $\times 10^4$ mu	Chemical atomic weight
Z	El.	A				
76	Os	190	26.4	190.038	2.0	193.1
		192	41.0	192.038	2.0	
77	Ir	191	38.5	191.040	2.1	
		193	61.5	193.941	2.1	
78	Pt	192	0.78			195.23
		194	32.8	194.040	2.0	
		195	33.7	195.040	2.0	
		196	25.4	196.039	2.0	
		198	7.23	198.044	2.2	
79	Au	197	100.	197.039	2.0	197.2
80	Hg	196	0.15			200.61
		198	10.12			
		199	17.04			
		200	23.25	200.028	1.4	
		201	13.18			
		202	29.54			
		204	6.72			
81	Tl	203	29.1	203.057	2.8	204.39
		205	70.9	205.057	2.8	
82	Pb	204	1.5	204.058	2.8	207.21
		206	23.6			
		207	22.6			
		208	52.3	208.057	2.7	
83	Bi	209	100.	209.055	2.6	209.00
84	Po					
85	At					
86	Rn	222*	.....	.....	.....	222
87	Fa					
88	Ra	223*	.....	.....	.....	226.05
		224*				
		226*				
89	Ac	227*	100.			232.12
90	Th	232*	100	232.12	5.2	
91	Pa	231*				
92	U	234*	0.006	.....	.....	
		235*	0.720			238.07
		238*	99.274	238.14	5.6	
93	Np	238*				
94	Pu	239*				
95	Am	241*				
96	Cm	242*				

TABLE 3. SPIN, MAGNETIC MOMENT, AND QUADRUPOLE MOMENT OF STABLE ISOTOPES

An asterisk \* indicates that the isotope is radioactive.

A dagger † indicates that the quadrupole moment is in megacycles.

The letters and numbers in brackets refer to the references given at the end of the table.

Isotope			Spin $i_0$ Units of $\hbar$	Magnetic moment, $\mu_0$ Units of $\hbar e/2Mc$	Quad. mom. $10^{-24}$ cm <sup>2</sup>	
Z	El.	A				
0	n	1	$\frac{1}{2}$	$-1.9103 \pm 0.0012$ [F3,P2,H7,A1,A3]	2.73	[K2]
1	H	1	$\frac{1}{2}$ [H8,K1]	2.7896 [K2,M15]		
		2	1 [M15]	$\mu^n/\mu^p = -0.6847 \pm 0.0004$ $0.85647 \pm 0.0003$ [A3,K3] $\mu^d/\mu^p = 0.30702 \pm 0.0001$ $\mu^p/\mu^d = 3.25719 \pm 0.00002$ [B7]		
2	He	3	$\frac{1}{2}$			
		4	0 [B5]	0.0		
3	Li	6	1 [M2]	0.8213 [R2,R3,M8]		
		7	$\frac{3}{2}$ [H2,G5,S6,F2]	3.2532 [R2,R3,M8,J4]		
4	Be	9	$\frac{3}{2}$	-1.176 [K13]		
5	B	10	1	$0.597 \pm 0.003$ [M9]		
		11	$\frac{3}{2}$	$2.686 \pm 0.005$ [M9]		
6	C	12	0 [B17]			
		13	$\frac{1}{2}$	$0.701 \pm 0.004$ [H3]		
7	N	14	1 [O1]	$0.402 \pm 0.002$ [M18,K14]		
		15	$\frac{1}{2}$ [K12,W3]	$-0.280 \pm 0.003$ [Z1]		
8	O	16	0 [B17]			
		17				
		18	0			
9	F	19	$\frac{1}{2}$ [G1,C1]	$2.622 \pm 0.014$ [R2,R3,M8]		
10	Ne	20	0			
		21				
		22	0			
11	Na	23	$\frac{3}{2}$ [J1,G3,E1,R1]	$2.216 \pm 0.011$ [K14]		
12	Mg	24	0			
		25				
		26	0			
13	Al	27	$\frac{5}{2}$ [H6,M10]	$3.628 \pm 0.010$ [M10]		
14	Si	28	0			
		29				
		30	0			
15	P	31	$\frac{1}{2}$ [J6,A2]	$\pm 1.1314 \pm 0.0013$ [P3]		
16	S	32	0 [B5]			
		33				
		34	0			
		36	0			
17	Cl	35	$\frac{3}{2}$ [T7]	$1.365 \pm 0.005$ [K15]	$-84 \pm 4^\dagger$ [T7]	
		37	$\frac{3}{2}$ [T7]	$-1.135 \pm 0.005$ [K15]	$-64 \pm 4^\dagger$ [T7]	
18	A	36	0			
		38	0			
		40	0			
19	K	39	$\frac{3}{2}$ [M6,F2]	$0.391 \pm 0.002$ [K14]		
		40*	4	-1.290		
		41	$\frac{3}{2}$ [M1]	$0.217 \pm 0.001$ [M1,K14]		
20	Ca	40	0			
		42	0			
		43	0			

TABLE 3. SPIN, MAGNETIC MOMENT, AND QUADRUPOLE MOMENT OF STABLE ISOTOPES—  
(Continued)

Isotope			Spin $i_0$ Units of $\hbar$	Magnetic moment, $\mu_0$ Units of $\hbar e/2Mc$	Quad. mom. $10^{-24}$ cm <sup>2</sup>
Z	El.	A			
20	Ca	44	0	4.8	[K10]
		46	0		
		48	0		
21	Sc	45	$\frac{7}{2}$ [S14,K7]		
22	Ti	46	0		
		47			
		48	0		
		49			
		50	0		
23	V	51	$\frac{7}{2}$ [K9]		
24	Cr	50	0		
		52	0		
		53			
		54	0		
25	Mn	55	$\frac{5}{2}$ [W2,F1]	3.0	[F1]
26	Fe	54	0		
		56	0		
		57			
		58	0		
27	Co	59	$\frac{7}{2}$ [K8,M11,R5]	2-3	[M11]
28	Ni	58	0		
		60	0		
		61			
		62	0		
		64	0		
29	Cu	63	$\frac{3}{2}$ [R8]	2.5 [S21,S26]	-0.1 $\pm$ 0.1 [S21]
		65	$\frac{3}{2}$ [R8]	-2.6 [S21,S26]	-0.1 $\pm$ 0.1 [S21]
				$\mu^{65}/\mu^{63} = 1.04$	
30	Zn	64	0		
		66	0		
		67	$\frac{5}{2}$ [L1]	0.9 [L1]	
		68	0		
		70	0		
31	Ga	69	$\frac{3}{2}$ [J1,C6]	2.0165 $\pm$ 0.0035 [P3]	0.20 [R6]
		71	$\frac{3}{2}$ [J1,C6]	2.5611 $\pm$ 0.0030 [P3]	0.13 [R6]
				$\mu^{71}/\mu^{69} = 1.270$	
32	Ge	70	0		
		72	0		
		73			
		74	0		
		76	0		
33	As	75	$\frac{3}{2}$ [T8,C7,C3]	1.5 [C3,S22]	0.3 [S22]
34	Se	74	0		
		76	0		
		77			
		78	0		
		80	0 [W5]		
		82	0		
35	Br	79	$\frac{3}{2}$ [T1,T7]	2.61 [T1,S2]	720 $\pm$ 10† [T7]
		81	$\frac{3}{2}$ [T1,T7]	2.61 [T1,S2]	556 $\pm$ 10† [T7]
				$\mu^{79}/\mu^{81} = 1.0$	

TABLE 3. SPIN, MAGNETIC MOMENT, AND QUADRUPOLE MOMENT OF STABLE ISOTOPES—  
(Continued)

Isotope			Spin $i_0$ Units of $\hbar$	Magnetic moment, $\mu_0$ Units of $\hbar e/2Mc$	Quad. mom. $10^{-24} \text{ cm}^2$
Z	El.	A			
36	Kr	78	0		
		80	0		
		82	0		
		83	$\frac{3}{2}$ [K11]	-0.967 [K17]	0.15 [K11,S26]
		84	0		
		86	0		
37	Rb	85	$\frac{3}{2}$ [K16,J8,M19]	$1.345 \pm 0.005$ [K14]	
		87*	$\frac{3}{2}$ [H10,M20,H9]	$2.741 \pm 0.009$ [K14]	
38	Sr	84	0		
		86	0		
		87	$\frac{3}{2}$ [H5]	-1.1 [H5]	
		88	0		
39	Y	89	$\frac{3}{2}$	> 0.1	
40	Zr	90	0		
		91			
		92	0		
		94	0		
		96	0		
41	Cb	93	$\frac{3}{2}$ [B6]	5.3 [M24]	
42	Mo	92	0		
		94	0		
		95	$\frac{1}{2}$		
		96	0		
		97	$\frac{1}{2}$		
		98	0		
		100	0		
43	Tc	99			
44	Ru	96	0		
		98	0		
		99			
		100	0		
		101			
		102	0		
		104	0		
45	Rh	103	$\frac{3}{2}$ [F5]		
46	Pd	102	0		
		104	0		
		105			
		106	0		
		108	0		
		110	0		
47	Ag	107	$\frac{3}{2}$ [J3]	-0.10 [J3]	
		109	$\frac{3}{2}$ [J3]	-0.19 [J3]	
				$\mu^{109}/\mu^{107} = 1.93$	
48	Cd	106	0		
		108	0		
		110	0		
		111	$\frac{3}{2}$ [S5,S7,S10]	-0.65 [J6,B5]	
		112	0		
		113	$\frac{3}{2}$ [S5,S7,S10]	-0.65 [J9,B5]	
				$\mu^{113}/\mu^{111} = 1.0$	

TABLE 3. SPIN, MAGNETIC MOMENT, AND QUADRUPOLE MOMENT OF STABLE ISOTOPES—  
(Continued)

Isotope			Spin $i_0$ Units of $\hbar$	Magnetic moment, $\mu_0$ Units of $\hbar e/2Mc$	Quad. mom. $10^{-24}$ cm <sup>2</sup>
Z	El.	A			
48	Cd	114	0		
		116	0		
49	In	113	$\frac{9}{2}$ [B1,M7]	6.4	
		115	$\frac{9}{2}$ [J2,P1,M7]	5.49	[B1,M7] [H1]
50	Sn	112	0		0.82 [B1,S24]
		114	0		
		115	$\frac{1}{2}$	-0.9	[T2]
		116	0		
		117	$\frac{1}{2}$ [T2,S11]	-0.89	[T2]
		118	0		
		119	$\frac{1}{2}$ [T2,S11]	-0.89	[T2]
		120	0		
		122	0		
		124	0		
51	Sb	121	$\frac{5}{2}$ [B3,T3,C3]	3.7	[C3,B5]
		123	$\frac{5}{2}$ [B3,C3]	2.8	[C3,B5]
				$\mu^{121}/\mu^{123} = 1.316$	
52	Te	120	0		
		122	0		
		123			
		124	0		
		125			
		126	0		
		128	0		
		130	0		
53	I	127	$\frac{5}{2}$ [T4,M13]	2.8122	[P3] -0.46 $\pm$ 0.15 [S3,S4,M14]
54	Xe	124	0		
		126	0		
		128	0		
		129	$\frac{1}{2}$ [K6,J7]	-0.9	[K6,J7,B5]
		130	0		
		131	$\frac{3}{2}$ [K6,J7]	0.8	[K6,J7,B5] 0 $\pm$ 0.1 [K11,S26]
				$\mu^{129}/\mu^{131} = -1.11$	
		132	0		
		134	0		
		136	0		
55	Cs	133	$\frac{7}{2}$ [K4,C8]	2.572 $\pm$ 0.013	[K14] < 0.3 [S6,K4]
56	Ba	130	0		
		132	0		
		134	0		
		135	$\frac{3}{2}$ [B4]	0.837 $\pm$ 0.003	[H4]
		136	0		
		137	$\frac{3}{2}$ [B4]	(0.936 $\pm$ 0.003)	[H4]
		138	0		
57	La	139	$\frac{7}{2}$ [A4]	2.5 - 2.8	[A5,C4,C5]
58	Ce	136	0		
		138	0		
		140	0		
		142	0		
59	Pr	141	$\frac{5}{2}$ [W1]		

TABLE 3. SPIN, MAGNETIC MOMENT, AND QUADRUPOLE MOMENT OF STABLE ISOTOPES—  
(Continued)

Isotope			Spin $i_0$ Units of $\hbar$	Magnetic moment, $\mu_0$ Units of $\hbar e/2Mc$	Quad. mom. $10^{-24}$ cm <sup>2</sup>
Z	El.	A			
60	Nd	142	0		
		143			
		144	0		
		145			
		146	0		
		148	0		
		150	0		
61*					
62	Sm	144	0		
		147			
		148*			
		149			
		150	0		
		152	0		
		154	0		
63	Eu	151	$\frac{5}{2}$ [S17]	3.4 [S17,S2]	$\sim 1.2$ [S17,C2]
		153	$\frac{5}{2}$ [S17]	1.5 [S17,S2]	$\sim 2.5$ [S26]
				$\mu^{151}/\mu^{153} = 2.24$	
64	Gd	152	0		
		154	0		
		155			
		156	0		
		157			
		158	0		
		160	0		
65	Tb	159	$\frac{3}{2}$ [S13]		
66	Dy	159			
		160	0		
		161			
		162	0		
		163			
		164	0		
67	Ho	165	$\frac{3}{2}$ [S18]		
68	Er	162	0		
		164	0		
		166	0		
		167			
		168	0		
		170	0		
69	Tm	169	$\frac{3}{2}$ [S15]		
70	Yb	168	0		
		170	0		
		171	$\frac{1}{2}$ [S26]	0.45 [S27]	
		172	0		
		173	$\frac{3}{2}$ [S26]	-0.65 [S27]	$3.9 \pm 0.4$ [S26]
				$\mu^{173}/\mu^{171} = 1.4$	
		174	0		
		176	0		
71	Lu	175	$\frac{3}{2}$ [S19,G2]	$2.6 \pm 0.5$ [G2]	5.9 [G2]
		176	$\geq \frac{1}{2}$ [H11,M21,L2,S28]	$3.8 \pm 0.7$ [S28]	6-8 [S28]

TABLE 3. SPIN, MAGNETIC MOMENT, AND QUADRUPOLE MOMENT OF STABLE ISOTOPES—  
(Continued)

Isotope			Spin $i_0$ Units of $\hbar$	Magnetic moment, $\mu_0$ Units of $\hbar e/2Mc$	Quad. mom. $10^{-24}$ cm <sup>2</sup>
Z	El.	A			
72	Hf	174	0		
		176	0		
		177	$\leq \frac{3}{2}$ [R4]		
		178	0		
		179	$\leq \frac{3}{2}$ [R4]		
		180	0		
73	Ta	181	$\frac{1}{2}$ [M22]	2.1	$\sim 6$ [S29]
74	W	180	0		
		182	0		
		183			
		184	0		
		186	0		
75	Re	185	$\frac{3}{2}$ [M4,M5,G4,Z2]	3.3 [S25,S26]	2.8 [S25,S26]
		187	$\frac{3}{2}$ [M4,M5,G4,Z2]	3.3 [S25,S26]	2.6 [S25,S26]
76	Os	184	0		
		186	0		
		187			
		188	0		
		189	$\frac{1}{2}$		
		190	0		
		192	0		
77	Ir	191	$\frac{1}{2}$ [V2]	$\mu^{191}/\mu^{193} = -1.0$ [V2]	
		193	$\frac{3}{2}$ [V2]		
78	Pt	192	0		
		194	0		
		195	$\frac{1}{2}$ [F4,V1,J5,T5]	0.6 [J5,T5,S1]	
		196	0		
		198	0		
79	Au	197	( $\frac{3}{2}$ ) [R7,W4]		
80	Hg	196	0	0.3 [R7,W4,S16,B5]	
		198	0		
		199	$\frac{1}{2}$ [S8,S9]	$0.547 \pm 0.002$ [M12]	
		200	0		
		201	$\frac{3}{2}$ [S8,S9]	$-0.607 \pm 0.003$ [M12] $\mu^{199}/\mu^{201} = -0.9018$ [S19]	0.5 [S30]
		202	0		
		204	0		
81	Tl	203	$\frac{1}{2}$ [M23,M17,S31]	1.45 [S24]	
		205	$\frac{1}{2}$ [M23,M17,S31]	1.45 [S24]	
82		204	0		
		206	0		
		207	$\frac{1}{2}$ [K5]	0.6 [M3,R9,S16,B5]	
		208	0		
83	Bi	209	$\frac{1}{2}$ [B2]	3.6 [S16,S26]	-0.39 [S20]

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## CHAPTER 2

### GAMMA RAYS

**2.1. Properties.** Gamma rays are an electromagnetic radiation produced only in nuclear processes, either in processes such as neutron capture or from the decay of excited nuclei by isomeric transitions. This distinguishes gamma radiation from x-rays only in the sense of its origin.

The energy of a gamma photon is directly proportional to the frequency:  $E = h\nu = hc/\lambda$ , where the factor of the proportionality,  $h$ , is Planck's constant,  $c$  is the velocity of light, and  $\lambda$  is the wavelength. In units of mev,  $E = 0.012354/\lambda$  where  $\lambda$  is in angstroms. Also associated with the photon is a momentum of magnitude  $p = h\nu/c = h/\lambda$ . In any process involving the interaction of a gamma photon with an atom or any elementary particle, both the energy and momentum must be distributed according to the laws of conservation among the various particles and radiations participating. In the collision of a photon with an atom, a large proportion of the momentum is transferred to the atom and the greater part of energy is transferred to an orbital electron which is ejected from the atom.

Gamma-ray absorption is due almost entirely to interaction of photons with free and bound electrons in an absorbing medium. Each absorption event, involving a single photon and an electron, takes place by one of three distinct interaction processes: (1) photoelectric effect, cross section designated by  $\tau$ ; (2) scattering (Compton effect), cross section designated by  $\sigma$ ; (3) pair formation, cross section designated by  $\kappa$ . The relative importance of each process as well as the absolute probability for its occurrence bears a strong dependence on gamma-ray energy and the atomic number of the absorber. In principle, the total effective electronic cross section over the entire energy range is represented by the sum of  $\tau$ ,  $\sigma$ , and  $\kappa$ , but in limited ranges contributions from more than one process may be negligible. At very low energies only the photoelectric effect is important, particularly in heavy elements. Scattering becomes the dominant process at medium energies, and for high energies pair formation is mainly responsible for gamma-ray absorption. Again, in certain portions of the energy range, pairs of processes, either  $\tau$  and  $\sigma$  or  $\sigma$  and  $\kappa$ , must be considered in gamma-ray absorption because the cross sections for the processes are then comparable in magnitude.

The simplest problem in gamma-ray absorption and the one most frequently encountered in experimental arrangements is that of a well-collimated beam of radiation. Geometrical reduction in intensity does not enter,

and photons scattered by the Compton effect are regarded as lost since they no longer contribute to the intensity of the collimated primary beam. Thus since each absorption event involves the complete removal of a single photon, the energy is not degraded as in the absorption of charged particles and the cross sections, therefore, remain constant. Consequently the reduction in energy flux, or intensity, is due wholly to the reduction in the number of photons and not the energy per photon. The rate at which the beam intensity or the number of photons decreases at any depth  $x$  in an absorber is then directly proportional to the depth.

$$\frac{dI}{dx} = -\mu x$$

Therefore if the absorber is homogeneous the gamma-ray intensity decreases exponentially with absorber thickness.

$$I = I_0 e^{-\mu x}$$

where  $I_0$  = intensity at surface

$\mu$  = absorption coefficient

The physical significance of the absorption coefficient is apparent from the fact that when  $x = 1/\mu$  the intensity is reduced to  $1/e$  of its value at the surface of the absorber. When  $x$  is measured in units of length,  $\mu$  is referred to as the linear-absorption coefficient and is expressed in  $\text{cm}^{-1}$ . Other units also can be used and sometimes are found more useful. A form that is frequently used is the mass-absorption coefficient  $\mu_m$  expressed in square centimeters per gram when  $x$  is measured in units of grams per square centimeter. This is related to the linear-absorption coefficient by  $\mu_m = \mu/\rho$ , where  $\rho$  is the density of the absorbing material. This form of the coefficient has the advantages of being an easy and more certain quantity to measure and is independent of the physical state of the absorber. Two other forms are also important in that they relate the linear and mass coefficients to values that can be calculated. The first is the electronic coefficient  $\mu_e$  which is simply the cross section per electron, or  $\mu_e = \tau_e + \sigma_e + \kappa_e$ . The second is the atomic-absorption coefficient or atomic cross section  $\mu_a$ , which is equal to  $\mu_e Z$ . The relationship between the four coefficients is then

$$\mu_l = \rho \mu_m = \rho N \mu_a = \rho N Z \mu_e$$

where  $N = N_0/A$  = number of atoms per gm

$N_0$  = Avogadro's number

$A$  = atomic weight

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where  $N = N_0/A$  = number of atoms per gm

$N_0$  = Avogadro's number

$A$  = atomic weight

$Z$  = atomic number

Instead of expressing gamma-ray absorption in terms of absorption coefficients it is sometimes more explicitly stated in units of half-value layers of

the particular absorber. One half-value layer is the quantity of material, either in grams per square centimeter or in centimeters, required to reduce the intensity of gamma rays of a particular energy to one-half its initial value at the surface. In terms of the absorption coefficient in corresponding units the half-value layer is  $T_{1/2} = 0.693/\mu$ . The gamma-ray intensity after traversing  $n$  half-value layers is then  $I = I_0 2^{-n}$ .

TABLE 4. MASS-ABSORPTION COEFFICIENTS FOR GAMMA RAYS

Data taken from "Handbook of Chemistry and Physics," 30th ed., Chemical Rubber Publishing Company, Cleveland, by permission of the publisher.

Energy, mev	Mass-absorption coefficients, cm <sup>2</sup> per gm							
	H	C	O	Al	Fe	Cu	Au	Pb
0.0296	0.390	0.256	0.372	1.170	8.45	11.45	28.4	32.0
0.0465	0.385	0.185	0.210	0.402	2.28	3.16	8.3	10.0
0.0617	0.375	0.175	0.183	0.270	1.10	1.55	4.40	4.90
0.0705	0.360	0.163	0.169	0.228	0.800	1.12	3.13	3.48
0.0845	0.340	0.155	0.162	0.195	0.520	0.680	7.85	2.35
0.0951	0.320	0.152	0.157	0.186	0.424	0.551	6.40	6.55
0.1260	0.280	0.142	0.144	0.156	0.265	0.325	3.21	3.50
0.1545	0.255	0.137	.....	0.146	0.235	0.268	2.42	2.50
0.1717	0.250	0.136	0.137	0.143	0.202	0.232	2.05	2.10
0.193	0.245	0.130	0.130	0.130	0.178	0.198	1.55	1.64
0.2470	.....	.....	.....	0.115	0.140	0.155	0.88	1.00
0.3085	0.205	0.110	.....	0.105	0.118	0.126	.....	0.62
0.4118	0.180	0.095	.....	0.093	0.095	0.100	.....	0.38
0.5147	0.165	0.080	.....	0.079	0.080	0.081	.....	0.21
1.2354	0.117	0.059	.....	0.058	0.058	0.057	.....	0.071
2.4708	0.078	0.0385	.....	0.038	.....	0.038	.....	0.042

It should be noted that the coefficients, cross sections, and half-value layers are constant only for a particular energy and absorbing atom. When the absorber consists of atoms with only one atomic number but the gamma radiation contains photons of several different energies, the intensity at any depth in an absorber is given by the sum of exponentials for the components.

$$I = I_1 e^{-\mu_1 x} + I_2 e^{-\mu_2 x} + \dots + I_n e^{-\mu_n x}$$

where  $I_1, I_2, \dots, I_n$  are the intensities of the  $s$  components at the surface and  $\mu_1, \mu_2, \dots, \mu_n$  are the corresponding absorption coefficients.

In practice, the experimental conditions involved in gamma-ray absorption are often more complicated: the energy may contain many components or may even be continuous in distribution, the absorber may contain several atomic species, and the geometry may be confused by a divergent beam.



These factors, together with the fact that scattering causes degradation of energy as well as directional divergence, make it impracticable to attempt to compute the reduction in energy flux or intensity of beams traversing absorbing media, and experimental measurements of the attenuation must be made. However, for the detection and measurement of gamma rays emitted by radioisotopes, the requirements of homogeneity and parallel beams can be met in the experimental arrangement and the simple exponential absorption described above is more nearly valid. Only scattered radiation remains a possible source of error, but this is largely eliminated by adequate collimation. Detailed calculations of gamma-ray absorption in thick slabs when single and multiple scattering are included have been given by Hirschfelder and Adams [13].

**2.2. Photoelectric Effect.** Gamma rays of low energy are absorbed mainly by photoelectric ejection of orbital electrons from atoms of the absorbing medium. This is a resonance phenomenon in which the energy of a photon is transferred to a single electron, ejecting it from the atom with a kinetic energy  $E_e$  equal to the difference between the gamma energy  $h\nu$  and the electron's ionization potential  $I$ , or  $E_e = h\nu - I$ . The mechanism of the interaction is most easily explained in terms of the influence of the electric and magnetic components of the gamma ray on an electron. At very low energies only the electric component is important, and since, as in other forms of radiation, it is oriented normally to the direction of the ray, the most probable direction taken by the ejected electron is also normal to the incident gamma ray and along the electric vector. As the energy is increased, the influence of the magnetic component becomes appreciable. The electron is accelerated by the electric field as before, but it is also deflected more in the forward direction of the incident photon by the magnetic-field component. From the direction taken by the electron it is evident that the total momentum involved in the process can be preserved only if it is shared in part with the atom from which the electron is ejected.

The orbital electrons most likely to participate in the photoelectric effect are those with binding energies nearest in magnitude to that of the incident photon, provided that the binding energy is less than  $h\nu$ . When the gamma-ray energy is considerably greater than the K x-ray absorption limit, the probability for interaction with electrons of the various atomic shells, K, L, M, . . . , decreases with increasing principal quantum number. Consequently the K electrons are the most likely to participate in the photoelectric effect when the gamma-ray energy is greater than the K absorption limit.

When the gamma-ray energy is comparable to the K, L, . . . electron ionization potentials, the photoelectric cross section is complicated by the characteristic x-ray absorption limits. However, for energies greater than

the K absorption limit the cross section, or absorption coefficient, decreases rapidly and smoothly. In this region the cross section per electron may be calculated from a formula given by Grey [13,15] in a form similar to that below.

$$\tau_e = 2.04 \times 10^{-30} Z^3 E_\gamma^{-4} (1 + 0.008Z) (E_\gamma - 0.25E_k - 0.422E_k^2)$$

where  $Z$  = atomic number of absorbing atoms

$E_\gamma$  = gamma-ray energy, mev

$E_k$  = K x-ray absorption limit, mev

When  $E_\gamma \gg E_k$ ,

$$\tau_e = 2.04 \times 10^{-30} Z^3 E_\gamma^{-3} (1 + 0.008Z)$$

From  $\tau_e$  the linear-, mass-, and atomic-absorption coefficients are obtained from the relations given in Sec. 2.1.

$$\tau_l = \rho \tau_m = \rho N \tau_a = \rho N Z \tau_e$$

When the electronic cross section is known for one substance, *e.g.*, lead, it may be calculated approximately for another absorbing material with atomic number  $Z$  by the relation

$$\tau_e = (\tau_e)_{\text{Pb}} \frac{Z^3}{(82)^3}$$

Similar extrapolations may be made for the linear and mass coefficients.

Empirical cross section formulas, equivalent to that above, have been given in the form

$$\tau_e = A Z^n \lambda^m$$

where  $A$  = constant

$\lambda$  = gamma wavelength

As an example, for air and water the mass-absorption coefficients are

$$\begin{aligned} &= 2.33\lambda^{3.13} && \text{(air)} \\ &= 2.54\lambda^{3.22} && \text{(water)} \end{aligned}$$

**2.3.\* Scattering of Gamma Rays.** Gamma rays are scattered with loss of energy only by electrons, and all electrons, whether bound in an atom or free, have the same cross section for scattering. It is the only process in which the gamma photon is not absorbed but instead undergoes a reduction in energy and a deflection from its initial direction. Thus a collimated beam of monoenergetic gamma rays after traversing an absorber is partially degraded in energy and spread over a wide angle by scattering.

Scattering is also the only process involving gamma rays in which both energy and momentum are balanced exclusively by the scattered photon and recoil electron; the atom does not participate in the interaction, and its presence is unnecessary. The effect was first described by Compton [1] who,

by considering the interaction as a classical two-body collision, derived the following relation between the scattering angle and change in wavelength (see Fig. 3):

$$\lambda' - \lambda = \frac{h}{m_0c} (1 - \cos \phi) = 0.0242(1 - \cos \phi) \quad \text{angstrom}$$

where  $\lambda'$ ,  $\lambda$  = initial and scattered wavelengths, angstroms

$\phi$  = scattering angle

The factor  $h/m_0c = 0.0242$  angstrom, known as the Compton wavelength, is the shift in wavelength for any gamma ray scattered through an angle of 90 deg. The scattering angle may take any value from 0 to  $\pi$ . Furthermore it is

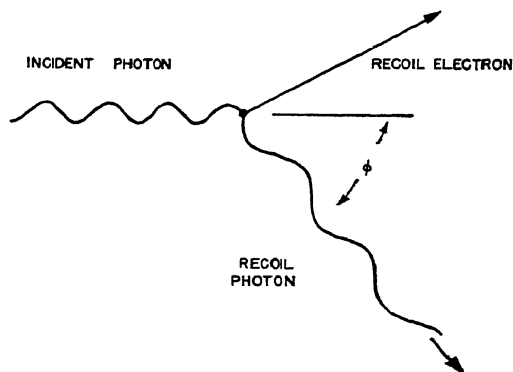


FIG. 3. Compton effect. A gamma photon is scattered by electrons as though it were a particle. The scattered photon consequently leaves with less energy (longer wavelength) and at a definite angle as required for conservation of energy and momentum in elastic, two-body collisions.

apparent that on the average after one or two collisions, high-energy gamma rays are degraded to wavelengths in the order of a Compton wavelength. Further scattering then has less marked effect, and the photon is more likely to be absorbed subsequently by the photoelectric effect.

For gamma-ray energies corresponding to the soft x-ray region, Compton scattering is negligible compared with the photoelectric effect, but at higher energies, approximately 0.5 to 5 mev, it is the most important process in gamma-ray absorption. For energies greater than this the scattering coefficient decreases slowly with energy and is rapidly superseded in importance by pair production. Furthermore, since the electronic cross section is independent of the atomic number of the absorber, scattering is relatively more important in light than in heavy elements as compared with the photoelectric effect and pair production, both of which exhibit strong dependence on  $Z$ .

A quantum mechanical treatment of scattering that is valid for all gamma-ray energies has been carried out by Klein and Nishina [2] on the basis of Dirac's relativistic theory of the electron [7]. The total electronic cross section is given as the sum of two scattering terms. The first, denoted by  $\sigma_e$ , accounts for the reduction in intensity due to loss of gamma photons scattered out of the beam. The second, denoted by  $\sigma_{se}$ , is the scattering-absorption cross section arising from the loss in energy suffered by scattered photons. The total electronic scattering cross section is given in the form [2]

$$\sigma_e = \frac{2\pi e^4}{m_0^2 c^4} \left[ \frac{2(1+\alpha)^2}{\alpha^2(1+2\alpha)} - \frac{1+\alpha}{\alpha^3} \log(1+2\alpha) - \frac{1+3\alpha}{(1+2\alpha)^2} + \frac{1}{2\alpha} \log(1+2\alpha) \right]$$

where  $\alpha = \frac{h\nu}{m_0 c^2} = \frac{E_\gamma}{0.51}$  = gamma-ray energy in units of  $m_0 c^2$  where  $E_\gamma$  is energy, mev

$e$  = electronic charge

$m_0$  = electronic rest mass

For very low and very high energies reduced expressions may be used. For low energies  $\alpha < 1$ ,  $\sigma_e$  can be expanded as [3]

$$\sigma_e = \frac{8\pi e^4}{3m_0^2 c^4} (1 - 2\alpha + 5.2\alpha^2 - 13.3\alpha^3 + 32.7\alpha^4 + \dots)$$

For high energies  $\alpha \gg 1$ ,  $\sigma_e$  reduces to

$$\sigma_e = \frac{2\pi e^4}{m_0^2 c^4} \left( \frac{1}{4\alpha} + \frac{1}{2\alpha} \log 2\alpha - \frac{1}{\alpha^2} \log 2\alpha \right)$$

The scattering-absorption coefficient  $\sigma_{se}$  is given by the equation [2]

$$\sigma_{se} = \frac{2\pi e^4}{m_0^2 c^4} \left[ \frac{2(1+\alpha)^2}{\alpha^2(1+2\alpha)^2} - \frac{1+3\alpha}{(1+2\alpha)^2} + \frac{(1+\alpha)(1+2\alpha-2\alpha^2)}{\alpha^2(1+2\alpha)^3} \right. \\ \left. - \frac{4\alpha^2}{3(1+2\alpha)^3} - \left( \frac{1+\alpha}{\alpha^3} - \frac{1}{2\alpha} + \frac{1}{2\alpha^3} \right) \log(1+2\alpha) \right]$$

As for  $\sigma_e$ , similar reduced expressions may be used for low and high gamma energies. For low energies  $\alpha < 1$ , and

$$\sigma_{se} = \frac{8\pi e^4}{3m_0^2 c^4} (\alpha - 4.2\alpha^2 + 14.7\alpha^3 - 46.17\alpha^4 + \dots)$$

For high energies  $\alpha \gg 1$ ,

$$\sigma_{se} = \frac{\pi e^4}{m_0^2 c^4} \left( \frac{1}{\alpha} \log 2\alpha - \frac{1}{3\alpha} \right)$$

The total electronic scattering cross section as indicated before is the sum

$\sigma_e = {}_s\sigma_e + {}_a\sigma_e$  from which the scattering coefficient  ${}_s\sigma_e$  can be obtained by subtraction.

The linear-, mass-, and atomic-absorption coefficients may be found from the usual ratios

$$\sigma_l = \rho\sigma_m = \rho N\sigma_a = \rho NZ\sigma_e$$

where  $\rho$  = density of absorber, gm per cc

$N = N_o/A$  = number of atoms per gm

$N_o$  = Avogadro's number

$A$  = atomic weight

$Z$  = atomic number

Since the value of  $\sigma_e$  is independent of atomic number, once it has been calculated for a desired energy range the curve may be applied to any absorbing material by multiplying it with the appropriate physical constants  $\rho$ ,  $N$ , and  $Z$ . Stated alternatively, the absorption coefficients are directly proportional to the electronic density of the medium.

**2.4. Pair Production.** Pair production involves the complete absorption of a gamma photon in the formation of an electron and a positron. The process can take place only in the coulomb field of a nucleus although the atom itself does not participate directly in the formation of the particles. Its presence is necessary mainly for the conservation of momentum when the gamma photon transfers its entire energy to the two light particles.

The cross section per electron for pair production increases very nearly in direct proportion to the atomic number of the absorbing atoms, but its variation with the gamma-ray energy is more complicated and computations with the exact formulas are inconvenient. It can be calculated, however, by approximations that are valid in particular energy ranges [4,5,6]. A special characteristic of pair production is the existence of a definite threshold energy at  $2m_0c^2$  (1.02 mev) below which the process cannot occur. Above the threshold the cross section increases rapidly for energies up to  $\sim 10$  mev, but at very high energies it increases more slowly, approximately as  $\log E_\gamma$ . An approximate formula for computing the cross section per electron for gamma-ray energies up to  $10m_0c^2$  has been given in a form similar to that below [13]

$$\kappa_e = 2.87 \times 10^{-28} Z(E_\gamma - 1.19)$$

where  $E_\gamma$  = gamma-ray energy, mev

For high gamma energies lying within the range where  $m_0c^2 \ll h\nu \ll 137 m_0c^2 Z^{-1/2}$ , Heitler and Sauter [16] and Bethe and Heitler [4] give an approximate formula for the electronic cross section in the form

$$\kappa_e = \left( \frac{e^2}{m_0c^2} \right)^2 \frac{Z}{137} \left( \frac{28}{9} \log \frac{2h\nu}{Z^{1/2}} - \frac{2}{27} \right)$$

where  $m_o$  = electronic rest mass

$e$  = electronic charge

$c$  = velocity of light

$Z$  = atomic number of absorber

At very great energies, where  $h\nu \gg 137m_o c^2 Z^{-1/2}$ , Bethe and Heitler [4] have also shown that the exact formula approaches asymptotically the value given by the formula

$$\kappa_e = \left( \frac{e^2}{m_o c^2} \right)^2 \frac{Z}{137} \left( \frac{28}{9} \log \frac{183}{Z^{1/2}} - \frac{2}{27} \right)$$

The atomic cross section and the linear- and mass-absorption coefficients for pair production are obtained from the usual relations

$$\kappa_l = \rho \kappa_m \quad \rho N \kappa_a = \rho N Z \kappa_o$$

When the mass-absorption coefficient is known for one substance, *e.g.*, lead, the coefficient for any other material may be found from

$$\kappa_m \quad (\kappa_m)_{\text{Pb}} \frac{207.2}{(82)^2 11.3} \frac{\rho Z^2}{A}$$

where  $\rho$ ,  $Z$ ,  $A$  = density, atomic number, and atomic weight, respectively, of absorber

$(\kappa_m)_{\text{Pb}}$  = mass-absorption coefficient for pair production in lead

The explanation of the process of pair production is to be found in Dirac's relativistic theory of the electron [7]. Dirac showed that the wave equation for the electron admits negative energy states for the electron as well as those of positive energy. The lowest positive state in the continuum of positive energies must necessarily be that equivalent to the rest mass of the electron,  $m_o c^2$ , as required by Einstein's law. Similarly, a highest negative energy state may exist at  $-m_o c^2$ , and below this there exists an infinity of possible quantum states identical to the positive energy continuum. Between  $-m_o c^2$  and  $+m_o c^2$  there are no states in which an electron can exist. Since the existence of electrons in positive states hardly needs demonstration, it must be assumed on the basis of Pauli's exclusion principle that all negative states are filled and, hence, that all space is occupied by an infinite density of negative energy electrons. The existence of such a "sea" of electrons normally could not be demonstrated because its uniformly distributed charge forms a field-free region. However if an electron is raised to a positive energy state, the unoccupied level or "hole" left behind behaves as a positively charged electron and can be detected by the ionization it produces. An electron can be ejected from a negative state only by the expenditure of energy at least equal to  $2m_o c^2$ , corresponding to the transition from  $-m_o c^2$  to  $+m_o c^2$ . Gamma rays with energies greater than this, therefore, can excite

an electron transition to positive energy states where it is then observed together with the "hole," or positron, as a pair of particles of identical mass but opposite charge. The energy in excess of  $2m_0c^2$  appears as kinetic energy, but it need not be shared equally by the two particles. When the positron is brought to rest by the normal processes of energy loss from ionization and radiation, it recombines with an electron, or more precisely, a positive energy electron fills the unoccupied level, and two gamma photons are emitted (annihilation radiation) each with a characteristic energy  $h\nu = m_0c^2$ .

The existence of such "holes" or positrons were first observed by Anderson [8] in cosmic radiation.

**2.5. Secondary Particle Production.** The total radiation observed at any depth in an absorber consists of primary gamma rays together with their

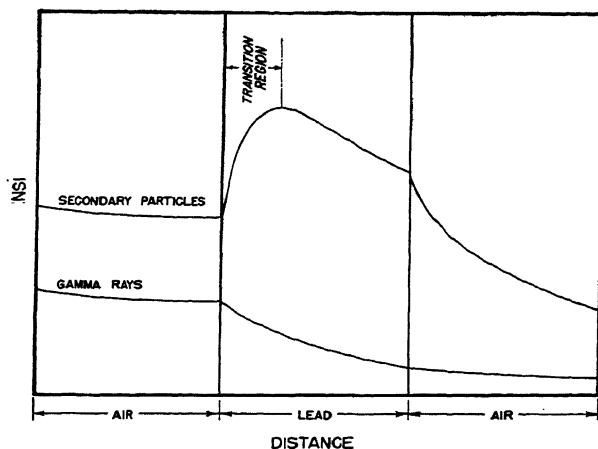


FIG. 4. Diagram indicating the change in secondary radiation intensity when gamma rays pass through media of different electronic densities—in this case, from air to lead to air. Intensity curves are not to scale.

secondary radiation of electrons (photo-, Compton, and pairs) and x-rays produced as a result of these electrons. Since the range of secondary electrons in all absorbers is very small compared with the half-value thickness for gamma rays, the number of recoil particles formed per unit time equals the number absorbed when radiative equilibrium is reached between the primary and secondary radiation intensity. The observed secondary intensity therefore decreases exponentially at the same rate as the gamma-ray intensity although its actual absorption coefficient is very much greater. The relative magnitudes of the intensity of the gamma-ray beam and its secondary electrons when they are in equilibrium depends on the atomic number and density

of the absorber, or more directly, on the separate absorption coefficients  $\mu$  and  $\mu_o$  as expressed by the relation

$$\frac{I}{I_o} = \frac{\mu_o}{\mu - \mu_o}$$

where  $I, I_o$  = intensities of secondary and primary radiations, respectively  
 $\mu, \mu_o$  = absorption coefficients of secondary and primary radiations, respectively

It is seen from the expression above that if the beam of gamma rays enters a second medium more dense than the first, *e.g.*, from air into lead, the intensity of the secondary radiation will make a transition to a higher relative intensity level before coming to equilibrium. The transition region is characterized by a very rapid increase in electron emission and reaches a maximum value at a depth in the absorber approximately equal to maximum range of the recoil electrons. From the surface of the second absorber (see Fig. 4) the variation in the secondary radiation intensity  $I$  with depth  $x$  is given by the equation

$$I = I_o \frac{\mu_o}{\mu - \mu_o} (e^{-\mu_o x} - e^{-\mu x})$$

where  $I_o$  = gamma-ray intensity at surface of absorber

$\mu_o$  = absorption coefficient for gamma ray

$\mu$  = absorption coefficient for secondary radiation

**2.6. Internal Conversion.** An isomeric transition in an excited nucleus is accompanied by the emission of either a gamma quantum or an electron. When a gamma quantum is emitted, it has its origin in the nucleus and possesses an energy  $h\nu$  equal to the difference in energy between the initial and final states of the nucleus. When an electron is sometimes found to accompany an isomeric transition, the gamma ray is said to undergo internal conversion. In such instances a gamma quantum is not emitted, but instead an electron is ejected from one of the innermost electron shells, K, L, . . . shells, of the same atom in which the isomeric transition occurs.

The kinetic energy of the conversion electron is exactly equal to the energy of the gamma ray with which it is associated minus the binding energy of the electron in the atom,  $E = h\nu - I$ . In general, for a large number of radioactive nuclei of the same species the conversion electrons associated with a particular isomeric transition are observed to fall into one or more monoenergetic groups with kinetic energies of  $E = h\nu - I_K$ ,  $E = h\nu - I_L$ ,  $E = h\nu - I_M$ , etc., where  $I_K, I_L, I_M, \dots$  are the ionization potentials of electrons in the K, L, M, . . . shells of the atom in which the transition occurs. It is important to note that if beta decay precedes the isomeric transition the binding energies to be used are those of the daughter isotope and not the parent.



Nuclei with complex decay schemes usually undergo several isomeric transitions to reach the ground state. Such multiple transitions may occur in cascade and as alternative branches, and each transition involves a different amount of energy. When gamma rays from several of these transitions undergo appreciable internal conversion, the energy spectrum of conversion electrons often exhibits considerable complexity since it may then consist of electron groups with kinetic energies determined by the various values of both  $h\nu$  and  $I$ . The relative intensities of the different groups show great variation. In most instances the strongest conversion occurs in the K shell followed by successively smaller contributions from the L, M, N, and, in a few cases such as ThC and RaB, the O shell. Among the lighter elements and for gamma-ray energies less than  $m_0c^2$ , the strongest conversion occurs in either the K or the L shell depending upon the energy and multipole order of the gamma quantum emitted in the transition. While K conversion is in most instances the strongest, L conversion in light elements,  $Z < 40$ , for gamma rays of low energy,  $\ll m_0c^2$ , and high multipole order tends to become more important. The ratio of K to L conversion under these conditions may have a value from 0.1 to  $\sim 10$  [18], as shown in Fig. 5.

The ratio  $\alpha = N_K/N_\gamma$ , the total number of conversion electrons divided by the number of gamma quanta associated with a given isomeric transition, is referred to in modern literature as the conversion coefficient. This form of the coefficient is to be preferred on physical grounds to the erroneous older expression  $N_e/(N_e + N_\gamma)$ , designated here by the letter  $f$ . Nevertheless, the value of  $f$  is still frequently used and in some instances is convenient since it expresses directly the fraction of transitions in which conversion electrons are emitted and the fraction  $1 - f$  in which gamma quanta are emitted. In complex decay schemes where branching occurs, the number of electrons and the number of gamma quanta emitted per disintegration depends also upon the transition probability  $p$  ( $\leq 1$ ) as given in the decay scheme. Hence the number of electrons per disintegration is  $fp$  or  $\alpha p/(\alpha + 1)$  and the number of quanta,  $(1 - f)p$  or  $p/(\alpha + 1)$ .

Theoretical investigations of internal conversion lead to a complicated

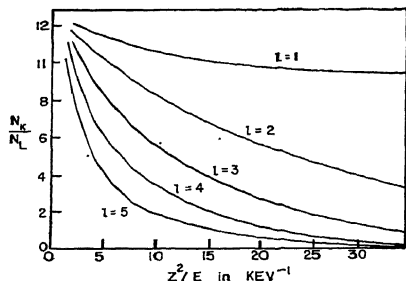


FIG. 5. Ratio of K to L conversion in elements for  $Z < 40$  and for  $E < M_0c^2$ .  $N_K$  and  $N_L$  are the numbers of K and L conversion electrons, respectively,  $l$  is the change in angular momentum of the nucleus in the transition, and  $E$  is the gamma-ray energy. [From M. H. Hebb and E. Nelson, *Phys. Rev.* **58**, 486 (1940).]

dependence of  $\alpha$  on the type of gamma radiation, which may be either electric or magnetic multipole, the multipole order of the radiation (dipole, quadrupole, . . .), the energy involved in the transition,  $h\nu$ , and the atomic number of the nucleus. For heavy nuclei and for gamma rays with  $h\nu > m_0c^2$ , calculations of  $\alpha$  [9,10,11] indicate stronger conversion for magnetic dipole than for electric dipole radiation and an increase in  $\alpha$  with multipole order. The most probable conversion under these conditions occurs in the K shell, while L, M, . . . conversion occurs with successively smaller probability for any one value of  $Z$  and  $E_\gamma$ . Conversion in lighter elements,  $Z < 40$ , and for low-energy isomeric transitions has been calculated for both the K shell [17] and the L shell [18]. The results of these investigations indicate that, for decreasing gamma-ray energy, conversion increases rapidly with multipole order. For any one energy and kind of radiation conversion varies approximately as  $Z^3$ .

Direct experimental evaluation of conversion coefficients usually has been obtained by beta spectrograph analysis of the beta spectrum. The line spectrum of monoenergetic conversion electrons appears as a set of peaks superimposed on the continuous background of the decay electrons when these are present. The peak positions in units of  $H\rho$  indicate the energies of electrons from various shells, and the area under the peaks indicate the relative intensities of the electrons. For a radioisotope with a simple decay scheme and one strongly converted gamma ray the conversion coefficient is given unambiguously by the ratio of the number of electrons in the line spectrum to the number in the continuous portion. However, when the decay scheme is complex, the method is difficult and the results often uncertain.

A second but less accurate method for determining conversion coefficients involves counter measurements of the coincidences ( $\beta e^-$ ) and ( $\beta\gamma$ ) [12,14]. A sample of the isotope for which internal conversion is to be measured is placed between two thin window counters one of which, counter *A*, registers single events as well as twofold coincidences with counter *B*. With sufficient aluminum absorber placed in front of counter *A* to stop all electrons, single gamma events are registered in *A* and ( $\beta\gamma$ ) coincidences by counters *B* and *A*. In the same way, but with the absorber removed, single beta events are now registered in counter *A* and ( $\beta e^-$ ) coincidences by counters *A* and *B*. The recorded number of ( $\beta e^-$ ) coincidences,  $N_{\beta e^-}$ , must be corrected for ( $\beta\gamma$ ) and ( $\gamma\gamma$ ) coincidences as measured above. These corrections are normally small, however, since the gamma-ray efficiency of counters is  $\sim 0.01$  whereas for beta particles the efficiency is  $\sim 1.0$ .

If the decay scheme of the isotope is simple and only one gamma is converted appreciably, the conversion coefficient is calculated from the counting measurements above by the expression

$$\frac{N_{\beta e^-}/N_{\beta}}{N_{\beta\gamma}/N_{\gamma}} \cdot \frac{2f}{\left(1 + f - \frac{f\epsilon\sigma}{2}\right)^2}$$

where  $f$  = total conversion coefficient

$N_{\beta e^-}$  = number of ( $\beta e^-$ ) coincidence counts

$N_{\beta}$  = number of beta counts

$N_{\beta\gamma}$  = number of ( $\beta\gamma$ ) coincidence counts

$N_{\gamma}$  = number of gamma counts

$\sigma$  = geometrical efficiency (per cent solid angle) of counter  $B$

$\epsilon$  = beta particle efficiency of counter  $B$ .

If  $f$  and  $\sigma$  are small, the term  $f\sigma\epsilon/2$  can be neglected. The ratio  $N_{\beta e^-}/N_{\beta}$  is in general a rapidly changing function of absorber thickness, the shape of the curve depending upon the relative values of the electron energy and the beta maximum energy. For this reason the effect of air and window absorption must be taken into account. In practice, the value of the ratio  $N_{\beta e^-}/N_{\beta}$  used to calculate  $f$  is determined by plotting the observed ratio as a function of absorber thickness and extrapolating, from the limiting value which the curve approaches, back to zero absorber.

If two cascade gamma rays are converted, the conversion coefficient determined by the above method is the sum of the two coefficients, but the separate values are not uniquely determined.

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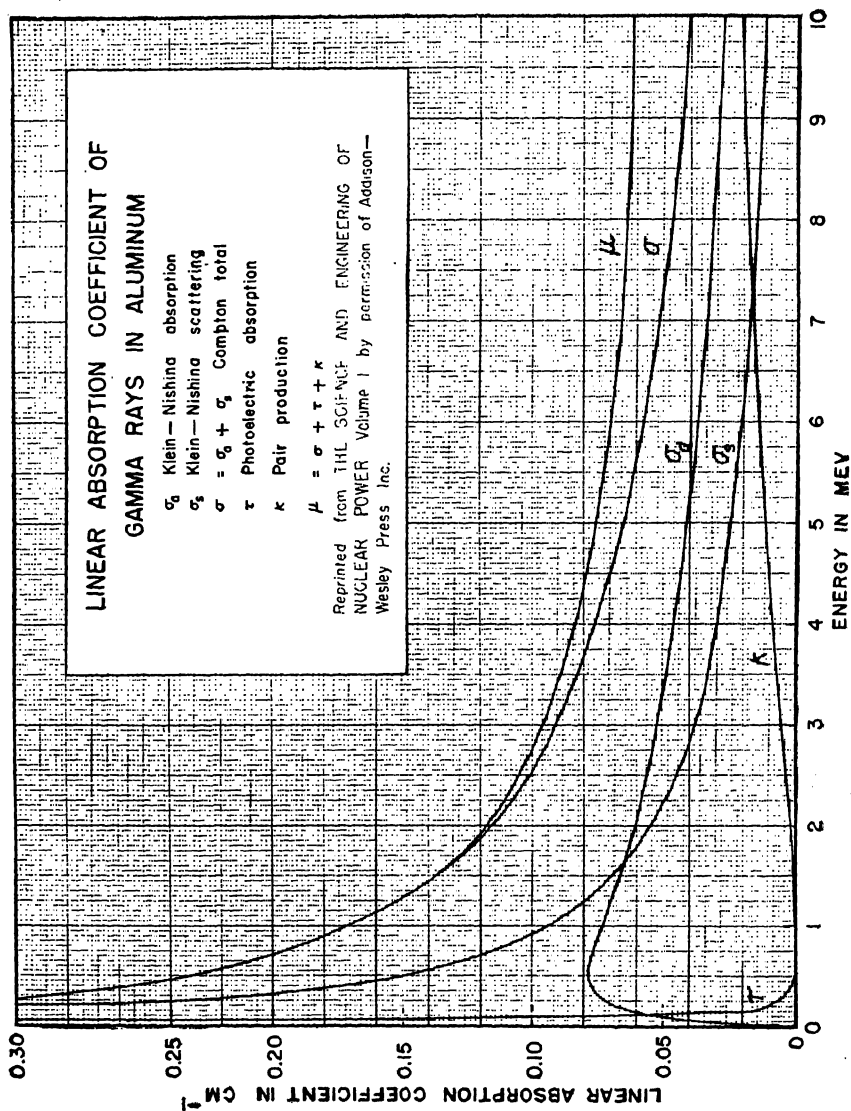
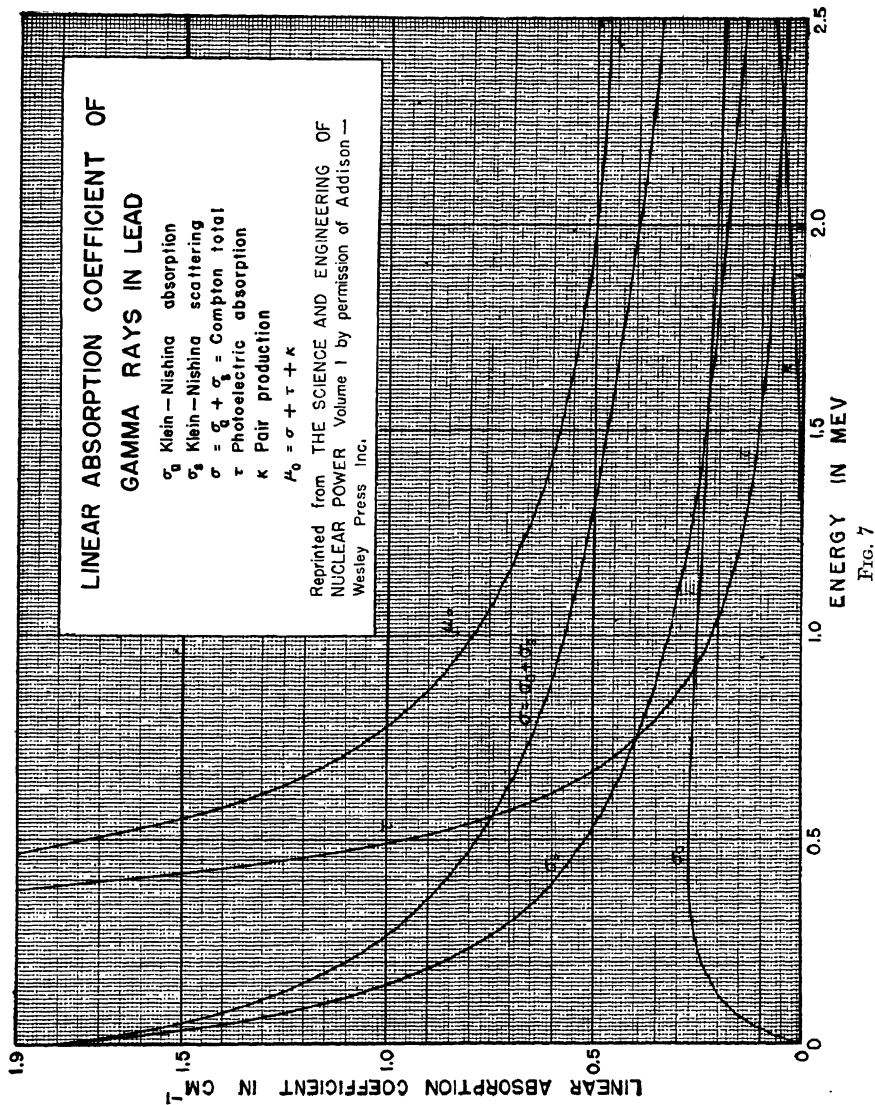


FIG. 6



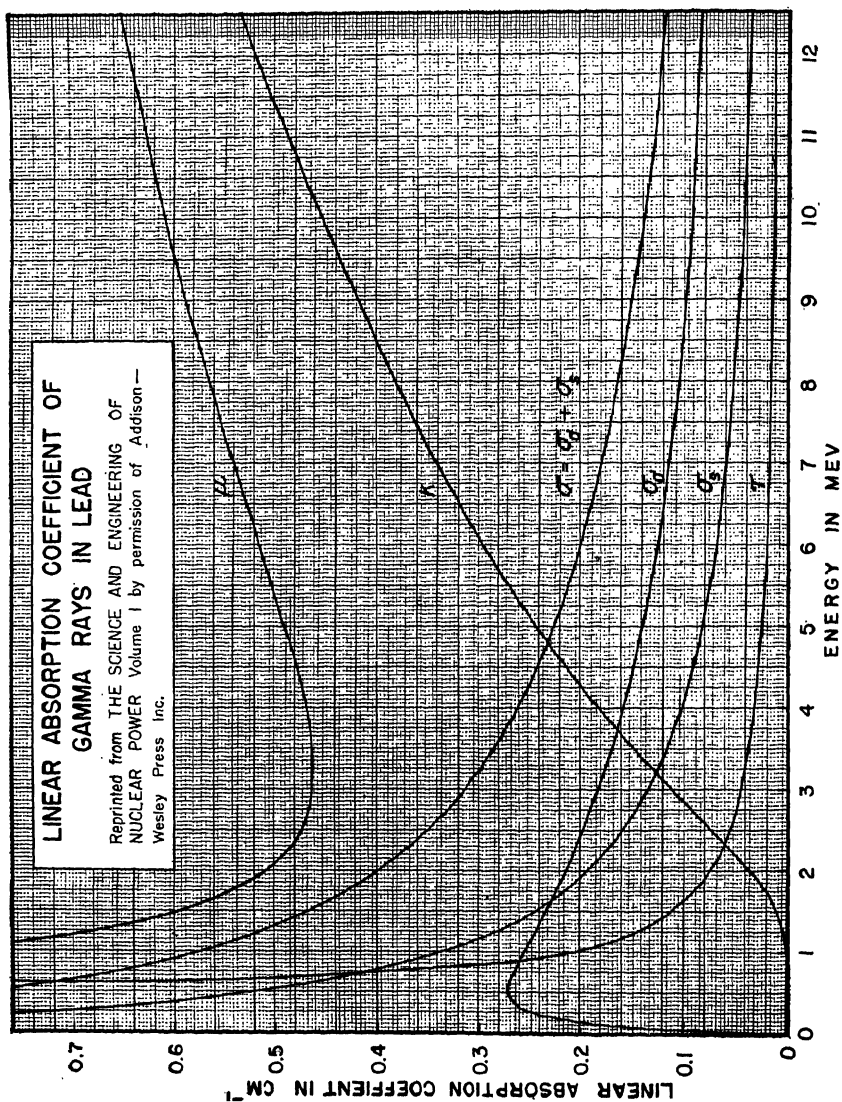


FIG. 8

## CHAPTER 3

### BETA PARTICLES

**3.1. Physical Properties.** Negatrons (negative electrons) and positrons (positive electrons) are identical in their interaction with matter and in their physical properties with the single exception of the sign of the charge associated with the particle. The following sections apply equally well to negatrons and positrons, observing only the proper sign of the charge where it occurs in formulas.

The static and dynamic properties of the beta particle are given below.

Rest mass [1]:	$m_o = (9.10660 \pm 0.0032) \times 10^{-28} \text{ gm}$
Charge [1]:	$e = (1.602033 \pm 0.00034) \times 10^{-20} \text{ abs emu}$ $= (4.80251 \pm 0.0010) \times 10^{-10} \text{ abs esu}$

Specific electronic charge [1]:	$\frac{e}{m_o} = (1.7502 \pm 0.0005) \times 10^8 \text{ abs emu/gm}$
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Relativistic mass:	$m = \frac{m_o}{\sqrt{1 - \frac{v^2}{c^2}}}$
--------------------	--

Momentum:	$p = \frac{m_o v}{\sqrt{1 - \frac{v^2}{c^2}}} = \frac{1}{c} \sqrt{T^2 + 2m_o c^2 T}$
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Kinetic energy:	$T = mc^2 - m_o c^2$ $= m_o c^2 \left( \frac{1}{\sqrt{1 - \frac{v^2}{c^2}}} - 1 \right)$
-----------------	--

where  $v$  = velocity of beta particle, cm per sec

$c$  = velocity of light, cm per sec

Energy is expressed usually in electron volts although units of  $H\rho$  are frequently used since this quantity can be measured directly with the beta spectrograph and in the Wilson cloud chamber operated in a uniform magnetic field. The relation between  $H\rho$  and energy is given by

$$H\rho = \frac{10^4}{3} \sqrt{E^2 + 1.02E}$$

where  $H$  = magnetic field strength, gauss

$\rho$  = radius of curvature, cm

$E$  = energy, mev

This relation is plotted in Fig. 9 for more convenient conversion.

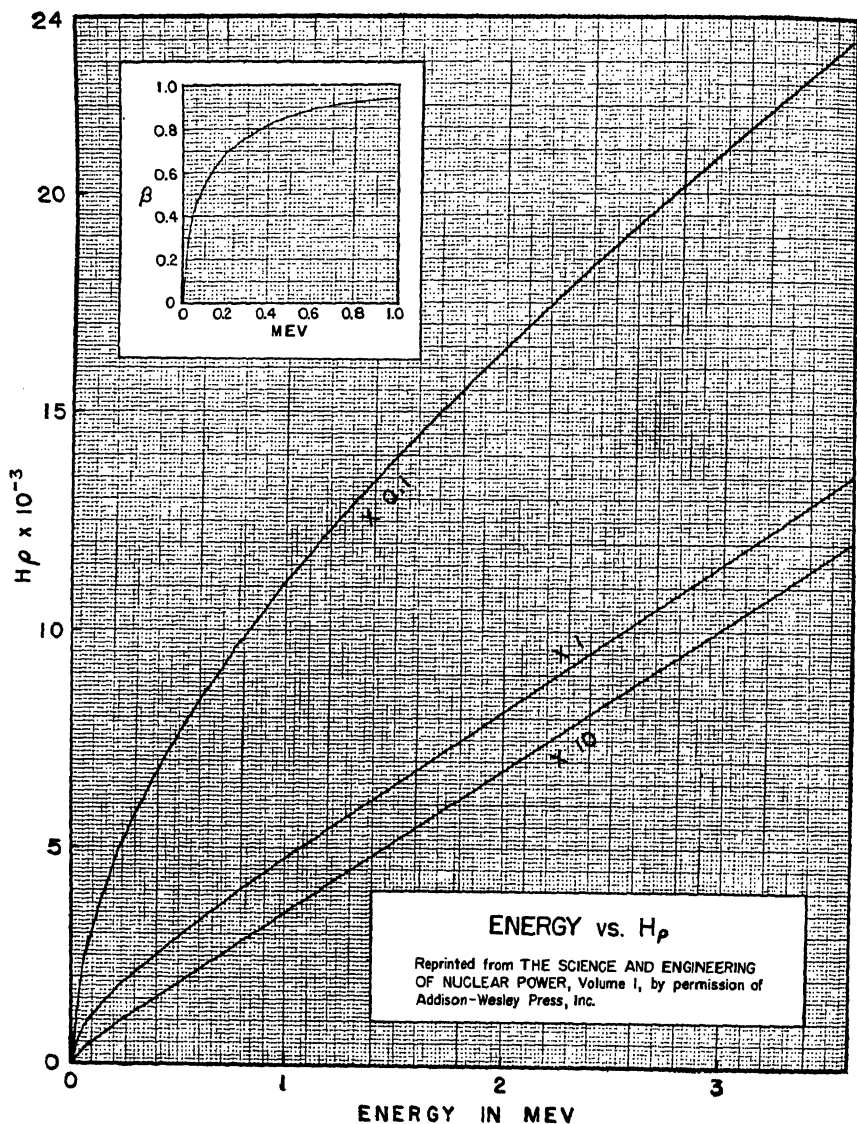


FIG. 9



**3.2. Absorption Processes.** The absorption of beta particles in matter is the result of energy loss by two main processes: excitation and ionization of atoms of the absorber and radiation of energy by the beta particle during acceleration in close collisions with nuclei. At sufficiently high energies ( $\sim 1$  mev or more) some loss occurs by nuclear excitation, but the contribution of this process to the stopping power of an absorber is wholly negligible compared with the two processes named above. At energies less than a few mev, radiative losses are also small and the principal stopping process is then excitation and ionization. At very high energies, however, radiative energy loss becomes dominant.

In principle, no clearly defined range-energy relation exists as it does for heavy charged particles because of the great variation in energy loss per collision, which may vary from zero to almost the total beta-particle energy, and because of the more pronounced effects of scattering. In practice, a maximum range can be established that is useful to dosimetry and for ascertaining the maximum energy associated with beta-particle beams.

**3.3. Ionization Energy Loss.** The mechanism of beta-particle energy loss from ionization and excitation of the absorber atoms follows in detail the corresponding processes for heavy charged particles. In brief, absorption of energy takes place by repeated transfer of fractions of the beta particle's kinetic energy to the orbital electrons through interaction between the field of the beta particle and that of the atomic electron. The stopping formula expressing the energy loss per unit length of path has been given by Bethe [2] in the form

$$-\frac{dE}{dx} = \frac{2\pi e^4 N Z}{m_e v^2} \left[ \log \frac{E^3}{m_e c^2 (1 - \beta^2) I^2} - \beta^2 \right]$$

where  $I$  = average excitation potential (see Alpha Particles, Chap. 4)

$Z$  = atomic number

$N$  = number of atoms per cc

$E$  = total energy of electron (kinetic plus rest mass)

$m_e$  = electronic mass

$e$  = electronic charge

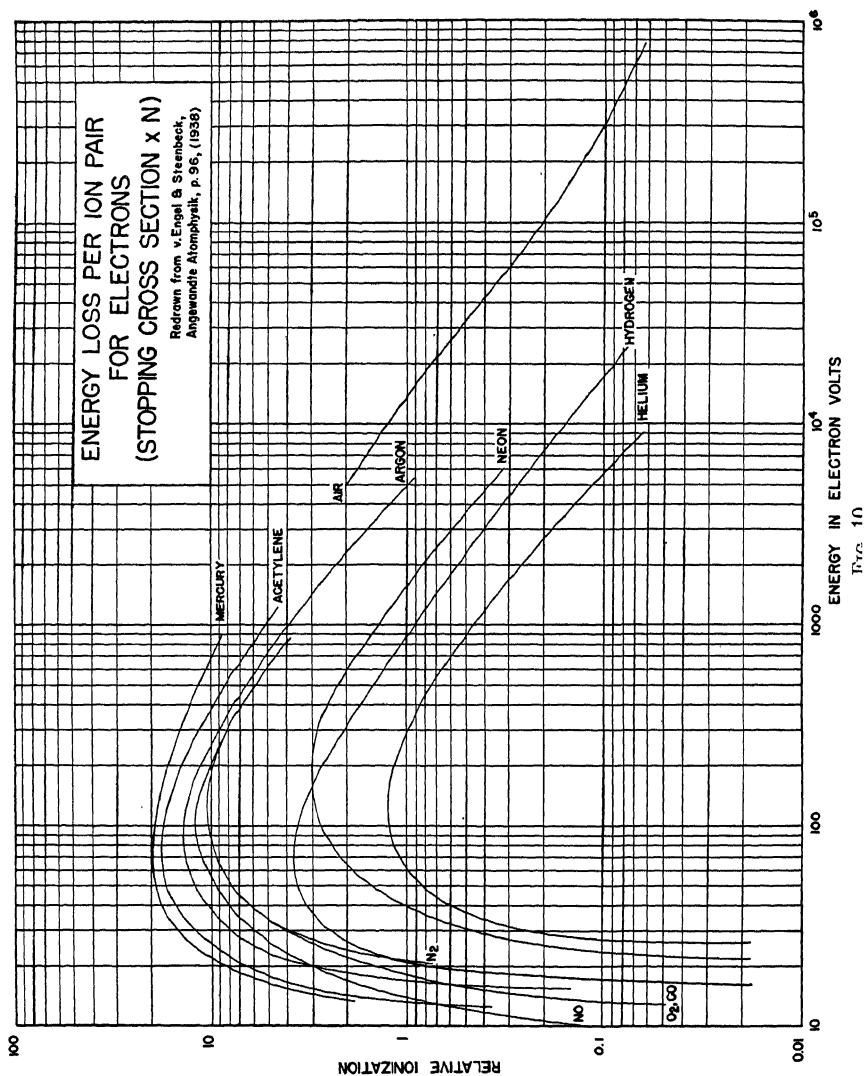
$v$  = velocity of electron

$\beta = v/c$

The stopping formula reduces to a simple form for electrons with very high energies,  $E \gg m_e c^2$ , traversing media of low density

$$-\frac{dE}{dx} = \frac{2\pi e^4 N Z}{m_e c^2} \log \frac{E^3}{2 m_e c^2 I^2}$$

This formula is not valid, however, for electrons with high velocities traversing dense materials since local polarization of the medium greatly alters the



field of the electron, and with it the rate of energy loss. Fermi's stopping formulas should be used when  $v \sim c\epsilon^{-1/2}$ , where  $\epsilon$  is the dielectric constant of the absorber (see Sec. 4.4).

Although these formulas may account adequately for the average energy loss by ionization, in practice they are of little use in calculating the actual absorption and range of beta particles since other factors also strongly influence absorption. (1) Unlike heavy charged particles, an electron may lose a large fraction of its energy in a single collision, and hence straggling occurs over a large portion of the range. (2) Beta particles are repeatedly scattered, often into large angles, making it impossible to correlate their actual path length with linear absorber thickness and also enhancing the straggling in any given direction. (3) Radiative loss, which is not taken into account in the stopping formula, increases with energy and becomes, at high energies, the more important factor in energy loss.

**3.4. Radiative Collision Losses.** At energies greater than several million electron volts radiative collisions account for an appreciable fraction of the energy loss. For this and higher energies, ionization losses increase very slowly, approximately as the log  $E$ , whereas radiative loss increases directly in proportion to  $E$ . At energies higher than 10 to 100 mev, depending on the atomic number of the absorber, radiation is the principal process for energy loss. Radiation emitted directly from beta particles traversing an absorber is known as *Bremsstrahlung* and is the source of the continuous or white x-ray spectrum.

The average energy lost per centimeter path length by radiation is given by an integral of the form

$$-\left(\frac{dE}{dx}\right)_{\text{rad}} = N \int_0^{v_e} h\nu\Phi_r d\nu$$

where  $N$  = number of atoms per cc

$\nu$  = frequency of radiation

The function  $\Phi_r$  has been derived by Bethe and Heitler [3], but in its general form, which includes the effect of screening by the outer orbital electrons, the function can be integrated only numerically. In two special cases of physical interest, however, the function reduces to integrable expressions and leads to the following formulas for calculating the average rate of energy loss by radiation:

For  $m_0c^2 \ll E \ll 137m_0c^2Z^{-1/2}$  (no screening),

$$-\left(\frac{dE}{dx}\right)_{\text{rad}} = \frac{NZ^2}{137} \left(\frac{e^2}{m_0c^2}\right)^2 E \left(4 \log \frac{2E}{m_0c^2} - \frac{4}{3}\right)$$

For  $E \gg 137m_0c^2^{-1/2}$  (complete screening)

$$-\left(\frac{dE}{dx}\right)_{\text{rad}} = \frac{NZ^2}{137} \left(\frac{e^2}{m_0c^2}\right)^2 E \left(4 \log 183Z^{-1/3} + \frac{2}{9}\right)$$

where  $Z$  = atomic number of absorber

$e$  = electronic charge

$m_0$  = electronic rest mass

$E$  = energy of electron in units of  $m_0c^2$

$c$  = velocity of light

In energy ranges other than those indicated above the complete expression for  $\Phi_r$  should be used and integrated numerically.

Neglecting the logarithmic terms in the stopping formula for ionization and in the formula above for radiative collisions, a simple but approximate formula is found for the ratio of the rates of energy loss by these two processes as a function of energy and atomic number.

$$\frac{(dE/dx)_{\text{ion}}}{(dE/dx)_{\text{rad}}} = \frac{EZ}{1,600 mc^2} = \frac{EZ}{800}$$

where  $E$  = energy, mev

A second form of radiative energy loss, known as Cerenkov radiation [20], occurs when high-speed electrons traverse dielectric media. Radiation is emitted in the frequency range for which the phase velocity in the medium is smaller than the velocity of the electron. The theory of the process, developed by Frank and Tamm [21], leads to expressions for the rate of emission given also by Fermi [28] in the form

For  $v < c\epsilon^{-1/2}$ ,

$$-\left(\frac{dE}{dx}\right)_{\text{Cer}} = \frac{2\pi ne^4}{mv^2} [-\beta^2 - \log(1 - \beta^2)]$$

For  $v > c\epsilon^{-1/2}$ ,

$$-\left(\frac{dE}{dx}\right)_{\text{Cer}} = \frac{2\pi ne^4}{mv^2} \left(\frac{1 - \beta^2}{\epsilon - 1} + \log \frac{\epsilon}{\epsilon - 1}\right)$$

where  $\epsilon$  = dielectric constant

$n$  = number of electrons per cc

$\beta = v/c$

In calculations of the total rate of energy loss from both ionization and radiation, the contribution from the Cerenkov effect given in the formula above should not be added since they are contained implicitly in Fermi's complete stopping formulas which should be used in the velocity range where the Cerenkov effect appears (see Sec. 4.4).

**3.5. Specific Ionization.** The total ionization produced by a beta particle is the sum of the primary ions produced directly by the particle plus the

subsequent ionization by the primary ions and that produced by *Bremsstrahlung* or x-rays emitted from the initial particle. In general, the range of the secondary ions is a fraction of a millimeter so that the majority of ions lie close to the path of the beta particle. The total ionization, however, usually amounts to at least several times the primary ionization produced directly by the initial particle. Differentiation between primary and secondary ionization is possible only in the Wilson cloud chamber with properly controlled expansion. When expansion takes place just before the electron traverses the chamber, the number of drops formed per unit length of path corresponds to the specific primary ionization. Under this condition secondary ions cannot diffuse sufficiently far to form separate drops but rather coalesce into a single drop containing the primary ion. If, on the other hand, expansion is delayed, the ions can diffuse far enough from one another to form separate drops and thus indicate quantitatively the total specific ionization.

The intensity of ionization is greatest (50 to 200 ion pairs per centimeter path in air) at very low beta particle velocities and decreases with greater velocity until a minimum value is reached when the energy is in the order of 1 mev. For energies greater than 1 mev, the specific ionization increases very slowly, roughly as  $\log E$ , but for practical purposes, in the energy range 1 to 10 mev, it remains essentially constant at approximately 25 ion pairs per centimeter path in air at normal temperature and pressure.

The average energy absorbed from a beta particle in the formation of an ion pair has been shown to be independent of velocity for energies greater than 0.01 mev. The value for air has been carefully measured, and its best value is given as 32.5 ev [22,23]. The energy absorbed per ion pair at low velocities is somewhat larger. For the energy range 300 to 60,000 ev G rbes [24] gives the following formula for the value of  $W$  in air:

$$W = 31.62 + \frac{5.27}{E - I} \quad \text{ev}$$

where  $E$  = beta-particle energy, kev

$I$  = ionization potential ( $1.7 \times 10^{-2}$  kev)

**3.6. Relative Stopping Power.** The relative stopping power  $S$  is defined as the ratio of the rate of energy loss in one substance to that in another substance, usually air. It is seen from the stopping formula for ionization energy loss that  $S$  should be relatively independent of velocity but strongly dependent upon the electronic density and to some extent on the average excitation potential of the medium. Considering only the stopping power per electron, the value of  $S_e$  in the range from hydrogen to copper decreases about 20 per cent [22] while, for any one substance, the variation with velocity is approximately 8 per cent in the energy range 0.1 to 2.0 mev [22].

**3.7. Nuclear Excitation.** Energy loss by nuclear excitation and capture is negligibly small in terms of absorption of beta-particle beams for all energies that have been observed. Its principal importance is in the determination of nuclear energy levels. Theoretical as well as experimental values of the cross section for nuclear excitation are of the order of  $10^{-7}$  barn.

**3.8. Absorption of Beta Particles of Homogeneous Energy.** A well-defined range does not exist for beta particles because of the great variation in energy loss per collision and, to an even greater degree, because of scattering. Nevertheless, a useful experimental range can be found since the absorption curve for a beam of initially monoenergetic beta particles such as conversion electrons is roughly linear for energies greater than 0.5 mev. Hence an extrapolated range may be determined by plotting the number of particles against thickness of absorber and extending the linear portion of the curve to the axis (or background count). Experimental determination of the range of particles with energies smaller than 0.5 mev by the absorption method is somewhat more difficult and less certain in its results than at higher energies. The absorption curve becomes more concave with decreasing energy and may, at low energies, be approximated by an exponential function over a considerable portion of the range. Estimation of the termination of the curve, however, is subject to considerable uncertainty, particularly if the background activity is appreciable. More accurate measurements of energy in this range are obtained with the beta spectrograph.

The relationship of energy to maximum range is also found to be linear for energies greater than 0.7 mev and at least as high as 3.0 mev. In this respect the range-energy relation for monoenergetic particles is almost identical to that for heteroenergetic beams, differing by only a few per cent for corresponding maximum energies. Figure 11 gives the observed extrapolated ranges of beta particles determined with the aid of the monoenergetic electrons provided by line spectra of various radioisotopes. An empirical relation for the extrapolated range in aluminum, fitting these data within  $\pm 5$  per cent over the energy range from 0.5 to 3 mev, is [4]

$$R = 0.52E - 0.9 \quad \text{mg/cm}^2$$

where  $E$  = energy, mev

The range in aluminum for energies less than 0.2 mev is given approximately by the relation proposed by Libby [26].

$$R = 1/150 E^{3/2} \quad \text{gm/cm}^2$$

**3.9. Absorption of Inhomogeneous Beta-particle Beams.** The absorption curve for beta particles of inhomogeneous energy decreases more rapidly than the curve observed for monoenergetic beams of the same maximum energy. The shape and range of such curves is of particular importance in its applica-

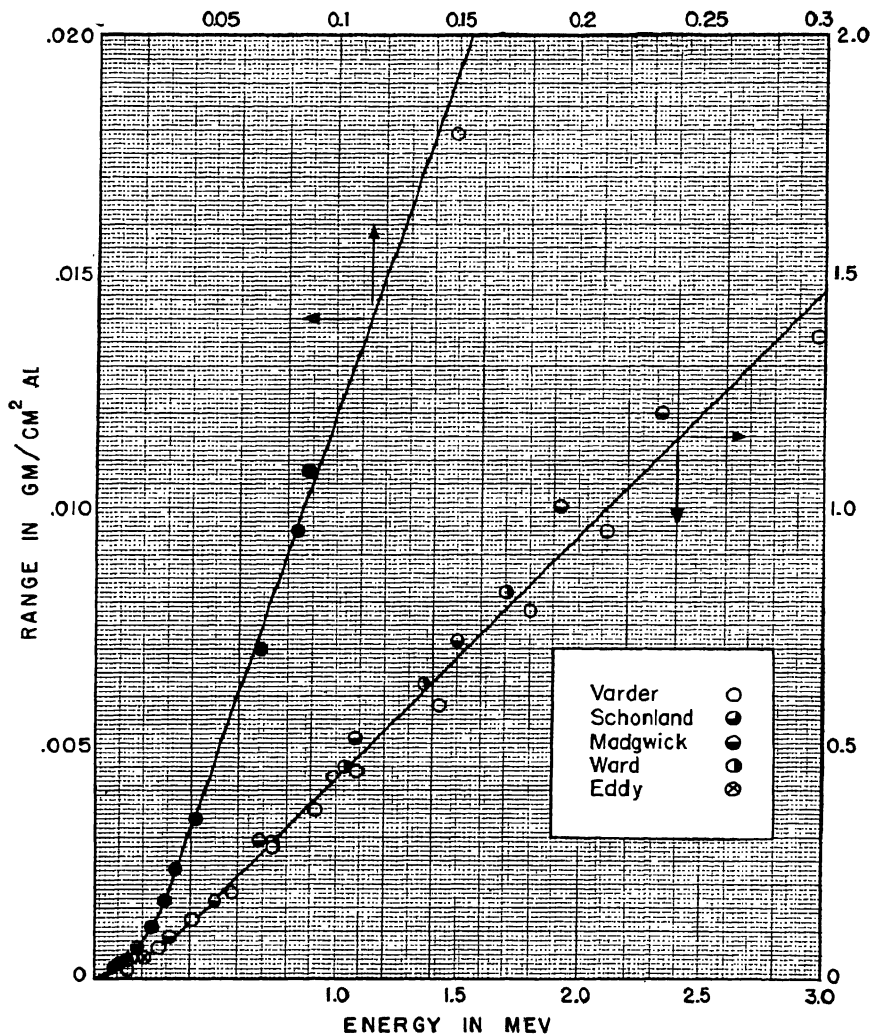


FIG. 11. Extrapolated range of monoenergetic beta particles in aluminum. (Reprinted from the "Science and Engineering of Nuclear Power," Vol. I, by permission of Addison-Wesley Press, Inc., Cambridge, Mass.)

tion to beta particles emitted from radioisotopes for which the energy distribution is continuous from zero to a well-defined maximum value. The observed absorption curve for beta particles from these sources is sometimes roughly logarithmic and often can be represented by an exponential function over the greater part of its range. Unlike similar curves for gamma rays, however, it has a finite and definite termination. The exponential character of the absorption can be interpreted only as a fortuitous effect due to a combination of the Fermi energy distribution of the emitted particles, scattering, and absorption by radiative and ionization energy loss. When the absorption curve does appear as a straight line when plotted on semilogarithmic graph paper, the beta intensity  $I$  (e.g., counts per minute) at a depth  $x$  in an absorber is given by

$$I = I_0 e^{-\mu x}$$

where  $I_0$  = initial intensity (or counts) with no absorber

$\mu$  = absorption coefficient, cm

$x$  = depth of absorber, cm

A form sometimes more convenient is found in terms of the mass-absorption coefficient  $\alpha = \mu/\rho$ , where  $\rho$  is the absorber density in grams per cubic centimeter. The absorber thickness  $x$  is then measured in grams per square centimeter and  $\alpha$  expressed in square centimeters per gram. This coefficient is relatively insensitive to the atomic number  $Z$  of the absorber since the number of electrons per unit mass decreases slowly with increasing atomic weight and for light elements is therefore essentially constant. An empirical relation for the mass-absorption coefficient, valid for the light elements and with a probable error in energy of 0.2 mev, is [4]

$$\alpha = \frac{22}{E_m^{1.33}}$$

where  $E_m$  = maximum energy, mev

and in terms of the half-value thickness  $d$  (where  $I = 0.5I_0$ ) the relation is

$$d = 0.693\alpha = 0.032E_m^{1.33}$$

These expressions are valid only when a considerable portion of the absorption curve appears, within the experimental error, as a straight line when plotted on semilogarithmic graph paper.

More often, when plotted on semilogarithmic graph paper with the absorber thickness as the linear abscissas, the absorption curve is found to be concave toward the origin or otherwise distorted and cannot be represented by a simple exponential function. The shape of the curve depends on the initial energy distribution of the beta particles and is also strongly influenced by the geometry of the counting arrangement. For this reason an absorption



coefficient is often of little use, and even when the absorption does appear to be exponential the counter geometry should be specified. A more definite quantity of universal adoption is the maximum range in aluminum. It is less subject to the influence of scattering and does not depend on the shape of the absorption curve.

Under certain conditions the range can be estimated from the plotted absorption curve, as shown in Fig. 12. The thickness of aluminum in milligrams per square centimeter just sufficient to stop all beta particles is the indicated range. From this value the true range is obtained by adding the equivalent absorber thickness of the air path between the source and the counter, the counter window, and the self-absorption in the source. When the absorber is a substance of low atomic weight, these corrections are given with sufficient accuracy by adding the products of the density of air, window, etc., and the path length in each. Normally when thin sources are used, the corrections do not total more than about 10 mg per  $\text{cm}^2$  equivalent of aluminum.

This method for measuring range is reliable only when the absorption curve can be followed through a reduction in intensity by a factor of 100 or more before reaching the level of background activity due to radiations other than the beta particles. Consequently the method cannot be used with accuracy for isotopes that also emit gamma rays. Their contribution to the counting rate, assuming a gamma-ray efficiency of  $\sim 1$  per cent, may amount to several per cent of the total, and since they are not appreciably attenuated by beta-particle absorbers, the gamma rays tend to mask the last portion of the beta absorption curve. Similarly when the source activity is less than several hundred times the background, it is often difficult to estimate with accuracy where the absorption curve terminates.

These difficulties are eliminated in a method of analysis developed by Feather [5] which is now generally adopted as the standard procedure for accurate range determinations. In addition to its accuracy as compared with other methods, it is applicable to complex as well as to simple spectra, to isotopes that emit gamma rays, and to relatively weak sources.

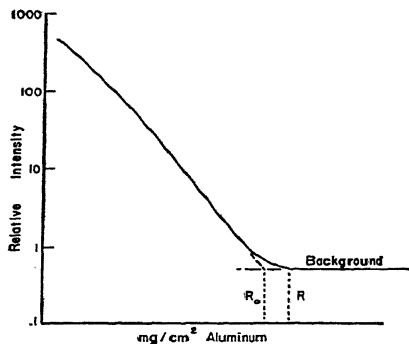


FIG. 12. Absorption curve of beta particles.  $R_0$  is the extrapolated range and  $R$  the maximum range. The absorption curve above is characteristic of the curves obtained for beta particles emitted from radioactive isotopes. For monoenergetic beta particles the relative intensity scale would be linear.

The Feather method consists in comparing the absorption curve of the beta emitter being investigated with the curve for an isotope with a simple beta spectrum for which the range is known accurately, such as RaE ( $476 \pm 2$  mg of aluminum per square centimeter [5]). The absorption curve for RaE, measured with aluminum-foil absorbers and corrected for air and window thickness and with the background subtracted, is plotted on semilogarithmic graph paper with range scaled along the abscissa. The total range (476 mg

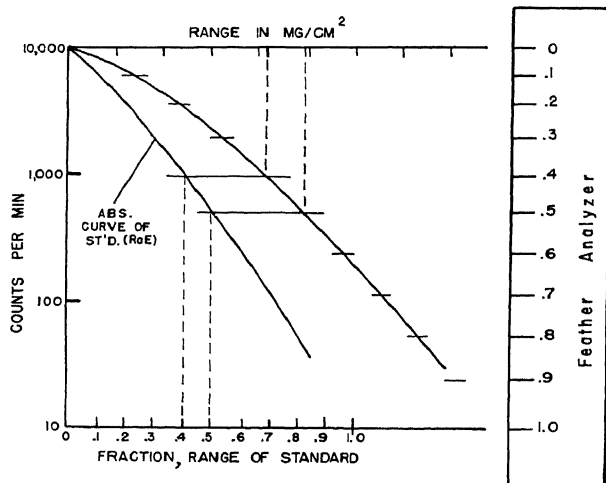


FIG. 13. Feather method for determining the range of beta particles by comparing the absorption curve with that of beta particles for which the range is accurately known (e.g., RaE for which  $R = 476$  mg/cm<sup>2</sup>).

per cm<sup>2</sup>) is then divided into 10 equal parts, and horizontal lines are drawn through corresponding points on the absorption curve, as shown in Fig. 13. Without altering the counter and sample geometry, a similarly corrected curve is obtained for the test material and plotted on the same graph starting also from the same ordinate for zero absorber thickness. The intersections of the previously drawn horizontal lines when referred to the abscissa give a set of fractional ranges differing from those of RaE. Each of the fractional ranges is then divided by its corresponding decimal fraction (0.1 — 0.9), and the resulting estimates of range are plotted against the same fraction, as illustrated by Fig. 14. A curve drawn through the points thus plotted may then be extrapolated to the point  $R$  (abscissa of 1.0) which is the actual range in aluminum. In practice the horizontal lines obtained with the RaE absorption curve are ruled on a vertical scale with the divisions marked 0 to 10 to match the corresponding fractional range. The scale may then be used

for any beta absorption curve which is measured with the same counting arrangement and plotted on the same kind of graph paper. The rest of the procedure is followed as before.

The Feather analysis can be extended to complex spectra provided that the difference in the maximum energies of the components is sufficiently great to allow the absorption curve to be broken down into components.

The relation between range and maximum energy of beta particles emitted from radioisotopes is linear within experimental errors for energies greater than 0.8 mev and at least as high as 3.0 mev. On the basis of range measurements made by the method above and corresponding energies determined with the beta spectrograph, Feather [5] proposed as the range-energy relation for beta particles in aluminum the expression

$$R = 543E - 160 \text{ mg/cm}^2$$

where  $E$  = energy, mev,  $> 0.7$

The constants were redetermined by Glendenin and Coryell [25] with more extensive data, and they give the relation as

$$R = 542E - 133 \text{ mg/cm}^2$$

For energies less than 0.8 mev the range-energy curve bends toward the origin in a way that cannot be represented accurately with a single function. For energies less than 0.2 mev, Libby [26] finds that the range is accurately represented by the formula

$$R = \frac{1}{150} E^{3/2} \text{ mg/cm}^2$$

In the range from 0.15 to 0.8 mev, the range is given by [12]

$$R = 407E^{1.38} \text{ mg/cm}^2$$

The portion of the curve for energies less than about 0.7 mev is essentially the same for monoenergetic and heteroenergetic beta-particle beams with the same maximum energy. The formulas above therefore can be used in either case.

**3.10. Scattering of Beta Particles by Nuclei.** The elastic scattering of beta particles has presented a difficult problem both in theory and in measurement. Because of the small mass of the electron compared to a nucleus,

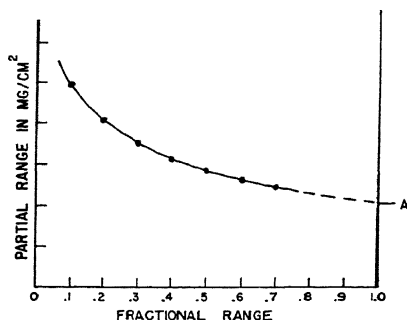


FIG. 14. Beta-particle range determination by the Feather method. The curve of partial ranges is extrapolated to full range (1.0) to obtain the actual beta-particle range given by the ordinate  $A$ .

large deflections in a single collision may occur frequently, particularly for beta particles of low energies scattered by heavy elements. For this reason, the effects of elastic scattering should never be underestimated in the evaluation of measurements on beta particles.

Single elastic collisions of slow electrons with nuclei can be calculated from Rutherford's formula [6] since elastic collisions involve only interaction of the coulomb fields of the electron and nucleus. For collisions of high-speed beta particles where relativistic effects are important, a formula has been given by Mott [7] for the intensity (or number) of beta particles scattered into the solid angle  $d\omega$  at an angle  $\theta$  from the initial direction.

$$n(\theta) = n_o N \left( \frac{e^2 Z}{2m_o v^2} \right)^2 (1 - \beta^2) \left[ \frac{1}{\sin^4 \theta/2} - \frac{\beta^2}{\sin^2 \theta/2} + \pi \beta \alpha Z \frac{\cos^2 (\theta/2)}{\sin^3 (\theta/2)} \right]$$

which for small angles and all values of  $Z$  reduces to

$$n = n_o N \left( \frac{e^2 Z}{2m_o v^2} \right)^2 (1 - \beta^2) \operatorname{cosec}^4 \frac{\theta}{2}$$

and for large angles and small  $Z$  is

$$n = n_o N \left( \frac{e^2 Z}{2m_o v^2} \right)^2 (1 - \beta^2) \left( 1 - \beta^2 \sin^2 \frac{\theta}{2} \right) \operatorname{cosec}^4 \frac{\theta}{2}$$

where  $\alpha = 2\pi e^2/hc = 1/137$

$n$  = number of particles per  $\text{cm}^2$  scattered into solid angle  $d\omega$  at angle  $\theta$

$n_o$  = initial number of particles per  $\text{cm}^2$  in beam

$N$  = number of atoms per cc

$Z$  = atomic number of scatterer

$m_o$  = electronic mass

In all cases the scattering coefficient is proportional to  $Z^2(1 - \beta^2)/v^2$ . These formulas apply only to very thin foils in which the probability of more than one collision is small. A criterion for determining if single scattering can be expected in an experimental arrangement has been given by Wentzel [8] which provides that the minimum observed angle of deflection  $\theta$  and the foil thickness  $t$  must be chosen so that  $\theta \geq 4\varphi$  where

$$\cot \frac{\varphi}{2} = \frac{m_o v^2}{e^2 Z} \sqrt{\frac{2}{\pi N t}}$$

where  $t$  = foil thickness, cm

This criterion permits several deflections through angles of the order of  $\varphi$  but determines the magnitudes of  $\theta_{\min}$  and  $t$  such that the contributions due to multiple collisions are negligible at an angle  $\geq \theta$  where the scattered beam is detected. For aluminum the maximum thickness for single scattering is

approximately  $2\mu$  (0.54 mg per  $\text{cm}^2$ ). In a more recent study of scattering, Williams [27] gives as the criterion for multiple scattering the expression

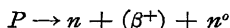
$$1 \ll \frac{nh^2 Z^{3/2}}{2\pi\beta m_e^2 c^2} ;$$

where  $t$  is in centimeters.

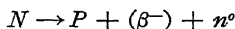
Single deflections of low-energy beta particles through large angles ( $> 90$  deg) may be expected with a finite but very small probability in "thin" foils but in "thick" layers where multiple scattering is important an appreciable fraction of an incident beam of particles is back-scattered, or deflected, through more than 90 deg. The coefficient of reflection  $R_s$  may be defined by the ratio  $I_r/I_o$  where  $I_o$  is the incident beta intensity and  $I_r$  is the back-scatter intensity. For most substances the reflection coefficient has a value between 0.1 and 0.5 depending upon the atomic number and density.

This effect is particularly important in measurements of radioactive samples and in the preparation of standards. The observed activity or counting rate is profoundly affected by the backing material. The back-scatter from platinum and lead, for example, is found to be 20 to 50 per cent greater than from aluminum, depending to some extent upon the counter geometry and beta-particle energy.

**3.11. Beta Decay.** The emission of beta particles (negatrons and positrons) occurs only during the decay of an unstable nucleus and is a slow process compared with the primary process in which an excited nucleus is formed by bombardment. The latter process takes place in less than  $10^{-10}$  sec, whereas the probable time for beta-particle emission is usually a second or longer. When an excited nucleus is formed by bombardment with heavy particles, it contains an excess of either protons or neutrons as compared with a stable nucleus of the same mass number. If the replacement of a proton with a neutron leads to a smaller atomic weight (greater binding energy), the transformation will occur according to the scheme



Conversely, if the replacement of a neutron with a proton results in a more stable nucleus, the transformation is



In both cases the excess energy (mass) is carried off by the neutrino  $\nu^0$  and beta particle. If a negatron is emitted, the nuclear charge increases by one unit; if a positron is emitted, the charge decreases by one unit. In calculating the exact mass reduction by these processes, the exact atomic weights (neutral atom) can be used in negatron emission since the loss of the rest mass of the beta particle is exactly compensated by the gain of an orbital electron. In

positron emission, however, a correction for twice the rest mass must be made since both the positron and an orbital electron are lost.

The emission of the hypothetical neutrino, first postulated by Pauli, is required for the conservation of energy, spin, and statistics. The emission of a beta particle corresponds to a transition in the excited nucleus involving a discrete quantity of energy; yet the emitted particles are observed to possess any energy ranging from zero up to the total transition energy,  $E_{\max}$ . It is necessary to assume, therefore, that the neutrino carries off an amount of energy in each instance equal to the difference between the beta-particle kinetic energy and the transition energy. Furthermore, beta emission always results in a change of spin  $i$  of the residual nucleus by integral units of  $\hbar/2\pi$ , *i.e.*, 0,  $\pm 1$ ,  $\pm 2$ ,  $\cdot \cdot \cdot$ , while the intrinsic spin of the beta particle is only a half unit. It must be assumed, therefore, that the neutrino spin is also one-half. A similar observation holds for the statistics in that residual nucleus remains unaltered while the emitted electron obeys Fermi statistics.

The properties of the neutrino deduced from these observations are then

Mass:	0 or $\ll$ electron mass
Charge:	None
Spin:	$\hbar/4\pi$
Magnetic moment:	$< 1.5 \times 10^{-4}$ Bohr magneton
Statistics:	Fermi-Dirac

With the aid of the neutrino hypothesis and the transformations ( $p \rightarrow n$ ), ( $n \rightarrow p$ ), Fermi [10] developed a theory of beta decay analogous to that for the emission of light from an atom. The theory provides a means of calculating the probability of the transformation in terms of the mean life for decay, the energy distribution of the particles and a set of selection rules for determining if a transition is allowed or forbidden.

Assuming the neutrino mass to be negligible, the probability of emitting per unit time a beta particle with energy between  $E$  and  $E + dE$  is given by Fermi's theory in the form

$$P dE = \frac{g^2}{2\pi^3} |Q|^2 f(Z, E) p E (E^2 - 1)^{1/2} (E_0 - E)^2 dE$$

where  $E$  = energy of electron in units of  $m_0c^2 = mc^2/m_0c^2$

$E_0$  = maximum energy in units of  $m_0c^2$

$g$  = constant

$p$  = momentum of electron

The factor  $Q$  is a matrix element involving the proper functions  $U_n$  for the neutron and  $U_m$  for the proton to which it is transformed, integrated over-all space and spin coordinates

$$Q = \int U_n^* U_m e^{-2\pi i(\mathbf{p}_n \cdot \mathbf{r} + \mathbf{p}_p \cdot \mathbf{r})/\hbar} d\mathbf{v}$$

If the proton or neutron that is transformed remains in nearly the same quantum state after beta emission,  $Q$  is nearly unity. If the initial and final quantum states are markedly different,  $Q$  is smaller than unity, and it is zero when a transition between two states is impossible; *i.e.*, the transformation of a proton or neutron in state  $n$  to a neutron or proton in state  $m$  is forbidden. For most light elements in which comparatively few states exist at ordinary levels of excitation, it can be regarded generally as unity (or zero if it is energetically impossible). The factor  $f(Z, E)$  includes the relativity correction and the appropriate wave function of the electron in the presence of the strong coulomb field of the nucleus.

In light elements  $f(Z, E)$  is of the order of unity; for  $Z = 0$ ,  $f(0, E) = 1$ . In heavy nuclei, however, this factor increases the probability for beta emission by an order of magnitude over that for light nuclei emitting beta particles of the same maximum energy.

The Fermi theory provides for a continuous distribution in kinetic energy for the ejected particles from zero to a maximum energy corresponding to the total transition energy for the particular nucleus and the specific transition. The most probable beta energy is found to be  $E_0/2$ , and near both zero kinetic and the maximum energy very few particles are to be expected. A difference in the numbers of positrons and beta particles should be found in the low-energy end of the spectrum for medium and heavy elements because of the strong effect of the nuclear coulomb field. Fewer low-energy positrons will be observed since, once formed, they derive additional energy from the repulsive electrostatic field. Conversely, a greater number of slow negative particles are observed because faster particles lose energy to the field and are then observed at relatively lower energy.

Measurements of beta-energy spectra are sometimes complicated in radioisotopes in which two or more beta particles are emitted or when conversion electrons (line spectra) are present (see Gamma Rays, Chap. 2). When a nucleus can decay by one of several possible beta transitions, the observed energy distribution is a superposition of the several Fermi curves [11]. Conversion electrons, on the other hand, can be readily identified since they form peaks with small energy spread and often high intensity superimposed on the continuous distributions.

An approximate expression for the energy distribution when  $E_0 < 2$  mev has been given in the form [12]

$$P dE = AE^{1/2}(1 + 2E)(1 + E)^{1/2}(E_0 - E)^2 dE$$

where  $A$  = constant for each isotope

A form more useful for plotting experimental data is to be found in the reduced equation given by Kurie [13].

$$\left(\frac{N}{\bar{F}}\right)^{1/2} = A - B(E + 1)$$

where  $N$  = number of particles observed in each successive momentum  
= interval

$E$  = energy in units of  $m_0c^2$

$A, B$  = experimental constants

In terms of the number  $N^1$  of particles per unit energy range, the "Kurie plot" is

$$\left(\frac{N^1}{pEF}\right)^{1/2} = C - DE$$

where  $p$  = momentum in units of  $m_0c$  for energy  $E$

$C, D$  = experimental constants

Plotting  $(N/F)^{1/2}$  or  $(N^1/pEF)^{1/2}$  against  $E$ , a straight line of negative slope is obtained if the particles follow a Fermi distribution in energy. The intercept on the energy ordinate corresponds to  $E_0 = \frac{1}{2}mv^2 + m_0c^2$ .

The theoretical mean life  $\tau$  for beta decay is found by integrating the probability of emission per unit time over the whole range of energy.

$$\frac{1}{\tau} = P_t = \int_1^{E_0} P dE = g^2 |Q|^2 F(Z, E_0)$$

where  $P_t$  = total probability that a particle of any energy is emitted per unit of time

$$F(Z, E_0) = \int_1^{E_0} E(E^2 - 1)^{1/2} (E - E_0)^2 f(Z, E) dE$$

The exact form of the function  $F(Z, E_0)$  is inconveniently complicated for calculation, but approximations valid over certain ranges of  $Z$  and  $E_0$  have been suggested [14,15,16]. In all cases the probable life for beta emission decreases rapidly with increasing maximum energy,  $E_0$ .

**3.12. Selection Rules for Beta Decay.** Nuclear transitions of the form  $n \rightarrow P$  or  $P \rightarrow n$  involved in beta emission are classed as allowed, first, second, or higher order forbidden depending upon the relative probability of the process. Empirically, a transition is classed by comparison of the magnitude of its half-life and energy with those of beta emitters of nearly the same atomic number.

The product of the half-life  $T$  and the function  $F$ , defined in the preceding section, is roughly constant for a particular class of transition within each of the three groups formed by light, medium, and heavy nuclei. Division of nuclei into at least three groups is necessary since the product  $FT$  increases also with atomic number independent of the forbiddenness of a transition.



This is due mainly to the more pronounced effect of the coulomb field with increasing  $Z$ . The orders of magnitude of  $FT$  for allowed, first and second forbidden transitions in light, medium, and heavy nuclei are given in Table 5 [15].

TABLE 5. VALUES OF THE PRODUCT  $FT$  IN BETA DECAY

Transition	$FT$		
	Light	Medium	Heavy
Allowed.....	$3 \times 10^3$	$5 \times 10^4$	$2 \times 10^5$
1st forbidden.....	$2 \times 10^5$	$2 \times 10^6$	$1 \times 10^7$
2d forbidden.....	$5 \times 10^7$	$1 \times 10^8$	$\sim 10^9$

Theoretical selection rules for beta decay are based on the form of the matrix element  $Q$  which determines the change in state accompanying beta emission. For many light beta emitters,  $Q \sim 1$  since the residual proton remains in nearly the same state as the initially transformed neutron. This is much less likely in heavy nuclei where the level density is much greater. If the change in state is considerable,  $Q \ll 1$  and the transition is to some degree, first, second, or higher order, forbidden. The principal changes in state to affect the probability of transition are the change in total angular momentum  $\Delta J$  of the nucleus and the parity change of the proper function. The selection rules originally proposed by Fermi are

Allowed:	$\Delta J = 0$	no parity change
1st forbidden:	$\Delta J = 0, \pm 1, (0 \rightarrow 0 \text{ forbidden})$	parity change
2d forbidden:	$\Delta J = \pm 1, \pm 2, (1 \leftrightarrow 0 \text{ forbidden})$	no parity change

A second set of selection rules proposed by Gamow and Teller [17] and based on a different choice of matrix element  $Q$  appears to find better support from the experimental data in certain instances. The simplest results of the G-T rules, without reference to the matrix elements, are

Allowed:	$\Delta J = 0, \pm 1, (0 \rightarrow 0 \text{ forbidden})$	no parity change
1st forbidden:	$\Delta J = 0, \pm 1, \pm 2, (0 \rightarrow 0 \text{ forbidden and } \frac{1}{2} \rightarrow \frac{1}{2} \text{ forbidden})$	parity change
2d forbidden:	$\Delta J = \pm 2, \pm 3, (0 \leftrightarrow 2 \text{ forbidden})$	$0 \rightarrow 0$ no parity change

**3.13. K Capture.** The inverse process to normal beta decay is the absorption by the nucleus of an orbital electron accompanied by the emission of a neutrino [18]. Once in the nucleus the electron does not retain its intrinsic form but is essential, as in positron emission, to the transformation of a proton to a neutron. The electron absorbed is one of the two K electrons since they

spend more time within and near the nucleus than do the L and M electrons and, hence, have a higher probability for capture when the process is energetically possible. K capture leads to a nucleus one charge unit less and to an atomic weight that is smaller than the initial atom by only the mass equivalence of the increase in binding energy.

The energy of the absorbed K electron has a definite value which is given by its rest energy minus its atomic binding energy, *i.e.*,  $E_o = (1 - \alpha^2 Z^2)^{1/2}$  in units of  $m_o c^2$ . Thus the neutrino must be ejected with a kinetic energy just equal to the sum of the total electron energy, and the nuclear transition energy and is therefore monoenergetic. Its value is given by

$$E_n = E_Z - E_{(Z-1)} + E_o - 1 = E_o + E_e$$

where  $E_o$  = transition energy in units of  $m_o c^2$

$E_Z$  = energy equivalent of exact atomic weight of initial nucleus in units of  $m_o c^2$

$E_{(Z-1)}$  = energy equivalent of exact atomic weight of final nucleus in units of  $m_o c^2$

Although K capture frequently competes with positron emission in the same transition, it is in some instances the only process energetically possible. Thus, only K capture is possible if the transition involves less energy than that equivalent to the rest mass of an electron (0.5 mev), *i.e.*, when

$$1 - E_o < E_Z - E_{(Z-1)} < 2 \quad (\text{units of } m_o c^2)$$

However, both positron emission and K capture are possible when  $E_o > 1$ .

The theoretical treatment and calculation of mean life and the transition probability is essentially the same as for beta decay. The probability of a transition depends principally, as in beta decay, on the accompanying change in total angular momentum  $\Delta J$  of the nucleus, *i.e.*,  $\Delta J = 0$ , allowed;  $\pm 1$ , first forbidden;  $\pm 2$ , second forbidden. The mean life is given by [15,19]

$$\frac{1}{\tau} = \frac{g^2}{2\pi^3} |Q|^2 f_k$$

where  $Q$  = matrix element ( $\sim 1$  for light elements; see Beta Decay, Sec. 3.11)

$g$  = constant

For allowed transitions [15],  $\Delta J = 0$ , the function  $f_k$  is approximately

$$f_k \approx 2\pi(\alpha Z)^3 (E_o + 1)^2$$

The probabilities of the first forbidden to the allowed transition will have the ratio  $(\alpha Z/2)^2$ , and successive higher orders of forbiddenness will have probability ratios of  $(E_o/R)^2$ , where  $\alpha$  is the fine structure constant,  $1/137$ , and  $R$  is the nuclear radius.

Observation of K capture is possible only through the detection of the K x-radiation following absorption of a K electron. However, identification of K capture is sometimes hampered by a competing positron emission when this is followed by gamma radiation. If internal conversion of the gamma radiation is pronounced, the K x-radiation is then masked by similar radiation produced by the conversion of K electrons. When positron emission is absent or when present but not accompanied by gamma radiation, K capture can be detected unequivocally. In some radioisotopes the gamma radiation exhibits a much greater intensity than is expected from the observed positron activity. This, together with K x-radiation, readily establishes the existence of K capture.

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TABLE 6. AVERAGE ENERGY OF BETA PARTICLES

The data given below are taken from the computed and compiled values of average beta energies reported by Marinelli, Brinckerhoff, and Hine, *Rev. Mod. Phys.*, **19**, 25 (1947). To ensure the highest accuracy they computed average values only from magnetic spectrometer measurements reported after 1939. In the case of complex spectra, the average value of the components, determined from graphical analysis, is given as well as the average for the total spectrum. The average for simple spectra was computed from Fermi's theory for the energy distribution by the formula

$$\bar{E} = \int_1^{E_0} EN dE / \int_1^{E_0} N dE \quad (\text{see Beta Decay, Sec. 3.11})$$

The column giving percentage indicates either the per cent of beta particles with different  $E_0$  or the per cent of positrons to K capture. In the case of positron emission the average total energy is indicated by the product of the particle average energy and emission probability per disintegration printed in parentheses.

The asterisk \* indicates that the values given refer only to the highest energy component of the spectrum whereas the dagger † indicates the lowest energy component.

Isotope			Rad.	Half-life, days	%	$E_0$ , kev $\pm$ kev		$\bar{E}$ , kev $\pm$ kev	$\bar{E}_t$ , kev $\pm$ kev	
Z	El.	A								
1	H	3	$\beta^-$	4,550.	100	17		.....	5.69	0.06
6	C	11	$\beta^+$	0.01415	....	970	10	.....	380	40
		14	$\beta^-$	$1.7 \times 10^6$	....	154		.....	50	
7	N	13	$\beta^+$	0.00703	....	1,240	20	.....	475	45
11	Na	22	$\beta^+$ , K, $\gamma$	1,170.0	$\sim 100$	575	30	.....	225	20
		24	$\beta^-, \gamma$	0.61	....	1,390	5	.....	540	20
15	P	32	$\beta^-$	14.5	....	1,712	8	.....	695	20
16	S	36	$\beta^-$	88.	....	167		.....	55	
17	Cl	38	$\beta^-, \gamma$	0.0259	53	4,940	60	2,230 90		
					11	2,790	60	1,190 40	1,390	70
					36	1,190	80	400 35		
19	K	40	$\beta^-, \gamma$	$5 \times 10^{11}$	....	1,350	50	.....	490	60
		42	$\beta^-, \gamma$	0.515	....	3,500		.....	1,395	
20	Ca	45	$\beta^-, (?)$	180.	....	260		.....	100	
21	Sc	44	$\beta^+, \gamma$	0.167	*	1,470	20	645 35		
		46	$\beta^-, \gamma$	85.					1,117	
23	V	48	$\beta^+, K, \gamma$	16.	58	715	15	.....	(300 $\pm$ 25)(0.58)	
25	Mn	52	$\beta^+, K, \gamma$	6.5	35	580	30	.....	(240 $\pm$ 20)(0.35)	
		56	$\beta^-, \gamma$	0.108	50	2,810	50	1,240 50	890	40
					30	1,040	30	410 35		
					20	650	100	280 25		
26	Fe	59	$\beta^-, \gamma$	47.	50	46	10	150 15		
						225	10	85 10	120	15
27	Co	55	$\beta^+, K$	0.75	....	1,500	50	.....	(515 $\pm$ 90)(?)	
		56	$\beta^+, \gamma$	85.	....	1,500	50	.....	655	35
		58	$\beta^+, K, \gamma$	65.	15	470	15	.....	(195 $\pm$ 20)(0.15)	
		60	$\beta^-, \gamma$	1,940.	....	310		.....	99	
29	Cu	61	$\beta^+, K, \gamma$	0.142	78	1,230	20	.....	(555 $\pm$ 40)(0.78)	
		64	$\beta^-$	0.53	....	578	3	175 30		
			$\beta^+, K, \gamma$	.....	30	659	3	265 25	(205 $\pm$ 30)(0.58)	
30	Zn	63	$\beta^+, K, \gamma$	0.0271	85	2,360	40	1,080 50		
					9	1,400	40	615 30	(985 $\pm$ 40)(0.98)	
					4	460	30	180 20		
33	As	76	$\beta^-, \gamma$	1.12	....	.....		.....	1,170	
35	Br	82	$\beta^-, \gamma$	1.5	....	465	10	.....	150	15
48	Cd	107	$\beta^+, K, \gamma$	0.28	0.31	320	10	.....	(140 $\pm$ 20)(0.003)	
									+92	
49	In	114	$\beta^-, (\gamma)$	50.	....	1,980	30	765 30	940	30

TABLE 6. AVERAGE ENERGY OF BETA PARTICLES—(Continued)

Isotope			Rad.	Half-life, days	%	$E_0$ , kev $\pm$ kev		$\bar{E}$ , kev $\pm$ kev		$\bar{E}_i$ , kev $\pm$ kev	
Z	El.	A									
51	Sb	124	$\beta^-$ , $\gamma$	60.	55	2,450	70	980	40	660	35
					45	740	30	260	25		
53	I	128	$\beta^-$ , $\gamma$	0.017		2,020		.....		770	
		130	$\beta^-$ , $\gamma$	0.525	45	1,030	20	360	20		
					55	610	30	195	20	270	20
		131	$\beta^-$ , $\gamma$	8.	....	595	10	.....		205	20
57	La	140	$\beta^-$ , $\gamma$	8.	12	2,120	80	835	60		
					60	1,400	40	510	40	495	40
					28	900	30	320	30		
77	Ir	194	$\beta^-$ , $\gamma$	0.81	*	218	40	.....		835	50
79	Au	198	$\beta^-$ , $\gamma$	2.7	....	970		.....		320	
83	RaE	210	$\beta^-$	4.85	....	1,170	5	.....		330	10
91	UX <sub>2</sub>	234	$\beta^-$ , $\gamma$	0.00079	*	2,320	5	.....		865	50
	UZ	234	$\beta^-$ , $\gamma$	0.28	90†	450	30	.....		150	20

TABLE 7. RANGES AND ABSORPTION COEFFICIENTS OF BETA PARTICLES FROM RADIO-ACTIVE ISOTOPEs

The data listed below are a compilation of values reported in recent literature. In order to avoid confusion in using the table, reference numbers for each value have been omitted but the principal literature from which data were obtained is listed at the end of the table. The maximum range, in mg/cm<sup>2</sup>, is given when known for aluminum and water as indicated by Al or W following its value. In most cases the range was determined by Feather's method. The absorption coefficient  $\alpha$  is defined by the usual expression  $I = I_0 e^{-\alpha t}$  where  $t$  is the thickness of aluminum absorber in milligrams per square centimeter. Values reported in the literature for the same isotope sometimes differ considerably in magnitude, but no effort was made to rectify these differences.

Isotope			Half-life	Energy, mev	Range, mg/cm <sup>2</sup>	$\alpha$ , cm <sup>2</sup> /mg
Z	El.	A				
1	H	3	12.5 y	0.017	0.23 Al	23
4	Be	10	$1.3 \times 10^8$ y	0.560	180 Al	
6	C	11		0.950	390 Al	
					415 W	
		14	4,700 y	0.140	20 Al	
					24 W	
7	N	13	10.13 m	1.20	550 W	
8	O	19	31 s	3.3	.....	2.56
9	F	18	112 m	0.700	260 Al	
		20	12 s	5.0	.....	0.93
11	Na	22	3.0 y	0.580	215 Al	
					210 W	
		24	14.8 h	1.40	620 Al	8.1
					640 W	
12	Mg	27	10.2 m	1.8	.....	2.56
13	Al	28	2.4 m	3.3	2,700 W	8.7
14	Si	31	170 m	1.80	860 Al	8
15	P	32	14.3 d	1.69	800 Al	
					810 W	5.3
16	S	35	87.1 d	0.107	13.5 Al	0.290
					20 W	
17	Cl	38	37 m	5.0	2,700 W	8.7
19	K	40	$4 \times 10^8$ y	0.40	610 W	
		42	12.4 h	1.52	~660 Al	} 2.56
				3.57	1,770 Al	
				3.57	1,900 W	
20	Ca	45	180 d	0.2	80 W	128
		49	30 m	.....	.....	7.5
21	Sc	44	3.92 h	1.5	680 W	~5.8
		46	85 d	0.26	100 W	7.1
22	Ti	51	6 m	1.6	.....	5.4
		51	72 d	0.36	.....	65
23	V	48	16 d	1.0	270 W	
		52	3.9 m	2.05	.....	3.9

TABLE 7. RANGES AND ABSORPTION COEFFICIENTS OF BETA PARTICLES FROM RADIO-ACTIVE ISOTOPES—(Continued)

Isotope			Half-life	Energy, mev	Range, mg/cm <sup>2</sup>		$\alpha$ , cm <sup>2</sup> /mg
Z	El.	A					
24	Cr	55	1.3 h	.....	.....		~8.3
25	Mn	52	6.5 d	0.77	220 W		
		56	2.59 h	(1.05, 2.86)	.....		4.85
26	Fe	59	47 d	0.26	150 W		43
27	Co	56	85 d	1.50	700 W		
		60	10.7 m	1.35	.....		147
		60	5.3 y	0.30	80 Al		79
28	Ni	65	2.6 h	1.9	.....		7.3
29	Cu	61	3.4 h	0.9	550 W		
		64	12.8 h	0.58 ( $\beta^-$ )	260 W		33
		66	5 m	2.9	.....		3.2
30	Zn	63	38 m	2.3	1,200 W		
		65	250 d	0.4	.....		107
		69	57 m	1.0	.....		17.1
		69	13.8 h	.....	.....		10.3
31	Ga	70	20 m	1.68	.....		10.3
		72	14.1 h	1.71	.....		9.6
32	Ge	71	40 h	1.2	.....		25.7
		75	89 m	1.1	.....		13.3
		77	12 h	1.9	.....		7.8
33	As	76	26.8 h	2.7	1,570 W		4.3
34	Se	81	19 m	1.5	.....		7.9
35	Br	80	18 m	2.0	.....		6.4
		82	34 h	0.465	160 W		35
37	Rb	86	19.5 d	1.6	.....		6.7
		88	17.5 m	4.6	.....		2.6
38	Sr	89	55 d	1.50	860 Al		8.8
					700 W		
	Sr	90	30 y	0.65	220 W		
39	Y	90	60 h	2.6	1,100 W		4.7
40	Zr	93	63 d	.....	.....		60
		95	17 h	1.0	.....		9.4
		97	6 m	2.0	.....		~5
41	Cb	94	6.6 m	1.4	.....		9.5
		95	35 d	0.15	25 Al		
42	Mo	99	67 h	1.5	520 Al		11.7
		101	19 m	1.8	.....		10
45	Rh	104	44 s	2.3	.....		3.9
		104	4.2 m	.....	.....		4.4
46	Pd	109	13 h	1.1	.....		16.1
		111	26 m	3.5	.....		6.9

TABLE 7. RANGES AND ABSORPTION COEFFICIENTS OF BETA PARTICLES FROM RADIO-ACTIVE ISOTOPES—(Continued)

Isotope			Half-life	Energy, mev	Range, mg/cm <sup>2</sup>	$\alpha$ , cm <sup>2</sup> /mg
Z	El.	A				
47	Ag	108	2.3 m	2.8	.....	6.7
		110	22 s	2.8	.....	2.9
		110	225 d	0.58	.....	36.5
48	Cd	115	43 d	1.5	840 Al	7.0
		115	2.5 d	1.1	.....	10.7
		117	3.75 h	1.3-1.7	.....	8.5
50	Sn	125	9 m	.....	.....	5.2
51	Sb	122	2.8 d	(0.8, 1.64)	.....	~ 10
		124	60 d	2.45	1,230 W	.....
52	Te	127	9.3 h	0.7	.....	27.2
		127	90 d	0.08	.....	~ 24
		129	72 m	1.8	.....	12.6
		131	25 m	.....	.....	7
		131	30 h	0.15	.....	~ 7.1
53	I	128	25 m	(2.02)	980 W	5.33
		130	0.525 d	0.61	450 W	.....
		131	8 d	0.6	210 Al	.....
56	Ba	139	86 m	1.	.....	7.9
		140	12.5 d	1.	390	.....
57	La	140	40 h	2.12	1,050 W	9.6
59	Pr	142	19.3 h	2.14	.....	5.4
62	Sm	153	46 h	0.7	.....	27.5
63	Eu	152	6.5 y	0.9	.....	26
		154	.....	.....	.....	.....
		155	15.4 d	.....	1,180 Al	.....
65	Tb	160	3.9 h	.....	.....	10
66	Dy	165	140 m	1.2	.....	12.4
		165	1.25 m	.....	.....	~ 50
67	Ho	166	30 h	1.9	.....	7.1
69	Tm	170	105 d	1.1	.....	17.3
71	Lu	176	3.5 h	0.215	48 Al	12.
		177	6.6 d	0.47	.....	50.
73	Ta	182	117 d	0.36	112 Al	46
74	W	185	77 d	0.72	.....	70
		187	24.1 h	0.5	.....	20.7
75	Re	186	90 h	1.07	.....	16.9
		188	18	2.05	.....	5.5
76	Os	191	30 h	(1.5, 0.95)	.....	14.5
		193	17 d	0.64	184 Al	.....
77	Ir	194	20.7 h	2.2	.....	6.1
79	Au	198	2.7	1.0	388 Al	19.3



TABLE 7. RANGES AND ABSORPTION COEFFICIENTS OF BETA PARTICLES FROM RADIO-ACTIVE ISOTOPES—(Continued)

Isotope			Half-life	Energy, mev	Range, mg/cm <sup>2</sup>	$\alpha$ , cm <sup>2</sup> /mg
Z	El.	A				
78	Pt	197	18 h	0.72	.....	26
		199	31 m	1.8	.....	9.4
80	Hg	203	51.5 d	0.11	15.3 Al	
				0.44	77.5 Al	9.5
		205	5.5 m	1.62	.....	9.4
81	Tl	204	4.23 m	1.77	.....	10
		206	3.5 y	0.87	.....	25.6
83	RaE	210	4.85 d	1.17	476 Al	
					520 W	16
91	UX <sub>2</sub>	234	1.14 m	2.32	1,105 Al	
					1,170 W	
	UZ	234	6.7 h	0.45	250 W	

## REFERENCES FOR TABLE 7

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## CHAPTER 4

### PROTONS, DEUTERONS, AND ALPHA PARTICLES

**4.1. Physical Properties.** The proton, deuteron, and alpha particle occupy a position of special importance. Aside from their significance as forms of the simplest and most accessible nuclear structures, they are the only charged heavy particles produced in nuclear reactions (except for the few cases in which  $H^3$  and fission fragments are formed) and the only heavy particles normally obtained from high-energy accelerators. Consequently the interaction of these particles with matter is of fundamental importance.

TABLE 8. PHYSICAL PROPERTIES OF THE PROTON, DEUTERON, AND ALPHA PARTICLE

Property	Proton	Deuteron	Alpha particle
Atomic weight (neutral).....	1.008123	2.014708	4.00390
Packing fraction, $10^{-4}$ mu.....	81.2	73.6	9.8
Charge, $10^{-20}$ abs emu.....	1.60203	1.60203	3.20406
Magnetic moment, nuclear magnetons.	2.7896	0.8565	
Binding energy, mev.....		2.17	28.
Spin, units of $\hbar/2\pi$ .....	$\frac{1}{2}$	1	0
Statistics.....	Fermi-Dirac	Bose-Einstein	Bose-Einstein

Qualitatively the interaction of protons, deuterons, and alpha particles with matter is very nearly the same for all three particles. Quantitatively the differences that exist are due almost entirely to the respective magnitudes of charge and mass of the particles. The following sections, with the exception of alpha decay, therefore, apply in principle equally well to all three particles. Physical theories of the structure of the particles and of their interaction with nuclei are rather involved and at the present time inconclusive because of the uncertainty of the nature of nuclear forces. Reviews of the various theories are to be found in the literature and will not be considered here.

**4.2. Energy Loss of Fast Charged Particles.** The mechanism chiefly responsible for the absorption or stopping of charged heavy particles ( $M \approx 1$  mass unit) is the interaction of their electric fields with the electrons of the absorbing material. The electrons are regarded, according to the classical theory of the process formulated by Bohr, as harmonic oscillators that are set in motion by the passing particle from which they derive energy. On the basis of this model the rate of energy loss is adequately accounted for by

the continual transfer of small fractions of the particle's kinetic energy to the excitation and ionization of atoms lying sufficiently near the path of the passing particle to be affected by its field. The greatest distance at which the field of the particle is effective in exciting atoms is of the order of  $2v/\nu(1 - \beta^2)^{1/2}$ , where  $v$  is the particle's velocity and  $\nu$  is the lowest vibrational frequency of the electrons. This corresponds to the radial distance beyond which the force on an electron due to the passing particle changes slowly compared to the electronic period in the atom. The interaction with distant atoms therefore may be treated by an adiabatic approximation which demonstrates that an electron, although temporarily perturbed, is left in its original state in an atom and does not absorb energy from the particle. A minimum radial distance from the path at which an atom can be excited is limited by the De Broglie wavelength  $\lambda = h(1 - \beta^2)^{1/2}/2\pi mv$ , since an approach closer than this has no significance. The relativistic factor  $(1 - \beta^2)^{1/2}$  is included to take into account the Lorentz contraction of the field at high velocities. In principle, the rate of energy loss is found by calculating the amount of energy transferred to electrons of all those atoms of the absorber lying within the cylindrical volume surrounding the path of the particle and defined by the maximum and minimum radii indicated above. This calculation has been subject to numerous theoretical studies which, though differing somewhat in detail, have led to formulas that are now highly satisfactory for computing the rate of energy loss and the range of heavy charged particles. The first derivation of a stopping formula was given by Bohr [24] and was subsequently developed mainly by Bloch [25] Bethe [4,5,6], Møller [7], Fermi [15], and Halpern and Hall [26].

Assuming that the stopping of charged particles results wholly from excitation and ionization, calculations of the rate of energy loss  $-dE/dx$  in simple substances compare remarkably well with values derived from accurately measured ranges of alpha particles emitted by the natural radioactive elements. The most commonly used and accepted expression at the present time probably is that derived by Bethe [4,5,6]. It is applicable to mesons, protons, deuterons, and alpha particles over a wide range of energy. In its simple form, including the relativity correction but excluding certain corrections discussed below, it is given as

$$-\frac{dE}{dx} = \frac{4\pi e^4 z^2 N}{mv^2} B$$

$$B = Z \left[ \log \frac{2mv^2}{I} - \log(1 - \beta^2) - \beta^2 \right]$$

where  $B$  = a dimensionless quantity called the stopping number

$e$  = electronic charge

$m$  = electronic mass

$N$  = number of atoms (not molecules) per cc

$z$  = charge number of particle

$Z$  = atomic number of absorbing atoms

$v$  = velocity of particle

$\beta = v/c$

$I$  = average excitation potential of atom

The range in energy over which the energy loss or stopping formula remains valid is determined (1) by the upper and lower limits inherent in its derivation and (2) by the appearance of effects at both low and very high energies which are not taken into account by the formula above.

The lower limit arises from the assumption that the particle's velocity is greater than the highest orbital electron velocity in an absorbing atom, i.e.,  $E \gg ME_K/m$ , where  $E_K$  is the K electron ionization potential and  $M$  and  $m$  are the masses of the particle and electron, respectively. This condition is necessary to ensure the constancy of the particle's charge  $ze$ . When the velocities of the incident particle and the electron are comparable, the probability of electron capture is no longer negligible and the charge of the particle becomes indeterminate since it then fluctuates with successive collisions. The probability of electron capture and loss as the particle is slowed down and brought to rest has not been completely formulated as yet, and only empirical corrections for its effect on the stopping formula are possible. This uncertainty in the charge leads to considerable error in attempting to calculate the rate of energy loss for protons with energies less than  $\sim 0.2$  mev and alpha particles with less than  $\sim 1.0$  mev. The same difficulty makes the stopping formula generally inapplicable to very heavy and multiply charged particles such as fission fragments since the charge varies more rapidly and in some unknown way with the velocity even at very high energies ( $> 100$  mev).

The highest energies for which the simple stopping formula above can be applied are limited by the appearance at very great energies of the Fermi effect described in the next section.

The average excitation potential  $I$  used in the stopping formula is usually considered to be a function only of the atomic number of the absorbing atoms and, in particular, to be independent of the kind of incident particle and its energy. As yet no completely satisfactory formula for its calculation has been provided. Its value can be determined empirically, however, with sufficient precision from accurately known range-energy data for protons or alpha particles in the absorber for which  $I$  is to be found. This is often done by adjusting the calculated range-energy curve to pass through the experimentally determined points by allowing  $I$  to be an arbitrary parameter [6,8,9,10]. The simplest and perhaps best theoretical value appears to be given by Bloch's conclusion [11] that  $I$  is directly proportional to the atomic number and by using Wilson's [30] empirical constant of proportionality, or

$I = 11.5 Z$  ev. The values for  $I$  for various substances determined empirically by Mano [12] and those calculated from the Bloch-Wilson formula are listed in Table 9. More recently Halpern and Hall [26] have introduced an analytical expression for  $I$  derived from a theory of ionization loss based on a multiple-dispersion-frequency model of the atom or molecule. In the energy range where the stopping formula above is accurate, the dispersion-frequency model gives the average excitation potential as the geometric mean of the dispersion frequencies  $\nu_i$  in the form

$$\log \nu_m = \sum_i^n f_i \log \nu_i$$

where  $f_i$  is the fraction of electrons with a dispersion frequency  $\nu_i$  and the sum is taken over all dispersion frequencies in the atom or molecule. The values of  $\nu_i$  must be obtained from spectroscopic data and are expressed in units of  $(N_e e^2 / \pi m)^{1/2}$  where  $N_e$  is the number of electrons per unit volume. The excitation potentials calculated by this method compare well with the values given in Table 9 only for light atoms; for heavy atoms they are as much as 20 per cent greater than the values given by Mano.

TABLE 9. AVERAGE EXCITATION POTENTIALS

Absorber	Z	I ev [Mano]	I ev (11.5 Z)	Absorber	Z	I ev [Mano]	I ev (11.5 Z)
$\frac{1}{2}\text{H}_2$	1	16.0	11.5	Cu	29	320	333.5
		17.5	23.0	Zn	30	340	345.0
He	2	34.5	.....	Kr	36	390	414.0
		44	.....	Mo	42	445	483.0
Li	3	38	34.5	Pd	46	490	529.0
$\frac{1}{2}\text{N}_2$	7	81	80.5	Ag	47	490	540.5
Air	7.23	87	83.15	Cd	48	495	552.0
		80.5 [6]	.....	Sn	50	500	575.0
		98.1 [13]	.....	Xe	54	530	621.0
$\frac{1}{2}\text{O}_2$	8	99	92.0	P	78	790	897.0
Ne	10	132	115.0	Au	79	780	908.5
Al	13	155	149.5	Pb	82	800	943.0
A	18	195	207.0				
Ni	28	325	322.0				

It is seen from the stopping formula that the rate of energy loss for particles of different mass but with the same velocity is proportional to the square of the particle's charge. Protons and deuterons consequently lose energy at one-fourth the rate of alpha particles that have the same velocity. This property of the stopping formula provides a convenient means for computing

energy loss and range data for various kinds of particles when detailed calculations of the stopping number have been made for a wide range in velocity for any one particle.

**4.3. Stopping-formula Corrections.** When applied to protons, deuterons, and alpha particles, the stopping formula given in the last section is reliable over a wide range of energy without further modification, but at low energies ( $< 10$  mev) and very high energies ( $> 1,000$  mev) corrections for more complicated effects must be inserted.

The largest error at very low energies is introduced by electron capture and loss, making the particles' time-averaged charge a function of velocity. The actual energy loss therefore is less than computed from the formula and for obvious physical reasons does not become infinite when  $v \rightarrow 0$  as indicated by the stopping formula. As yet, however, no general theoretical formulation of a correction for this effect has been given. The error becomes appreciable for alpha particles below 1.0 mev and for protons below approximately 0.2 mev.

A second correction, formulated by Bethe [6], for low-energy particles arises from the reduced contribution of the K electrons, and in principle, also the L and M electrons, to the stopping power of medium and heavy atoms due to the screening effect of the outer orbital electrons. The correction for the reduced stopping power of the K electrons is contained in modifications of the stopping number  $B$  which, as given by Bethe [6], have the forms

$$\text{For } E \leq \frac{M}{m} Z_{\text{eff}}^2 I_H,$$

$$B = (Z - 1.81) \log \frac{2mv^2}{I'} + B_K$$

$$\text{For } E \geq \frac{M}{m} Z_{\text{eff}}^2 I_H,$$

$$B = Z \log \frac{2mv^2}{I} - C_K$$

where  $E$  = energy of particle

$M$  = mass of particle

$m$  = electronic mass

$I_H$  = ionization potential of hydrogen atom, 13.60 ev

$I'$  = average excitation potential of electrons outside K shell

$I$  = average excitation potential of entire atom

$Z_{\text{eff}}$  = effective nuclear charge in K shell

The numbers  $B_K$  and  $C_K$  are plotted in Fig. 15 as functions of  $\eta$  where

$$\eta = \frac{mE}{MI_H Z_{\text{eff}}^2}$$

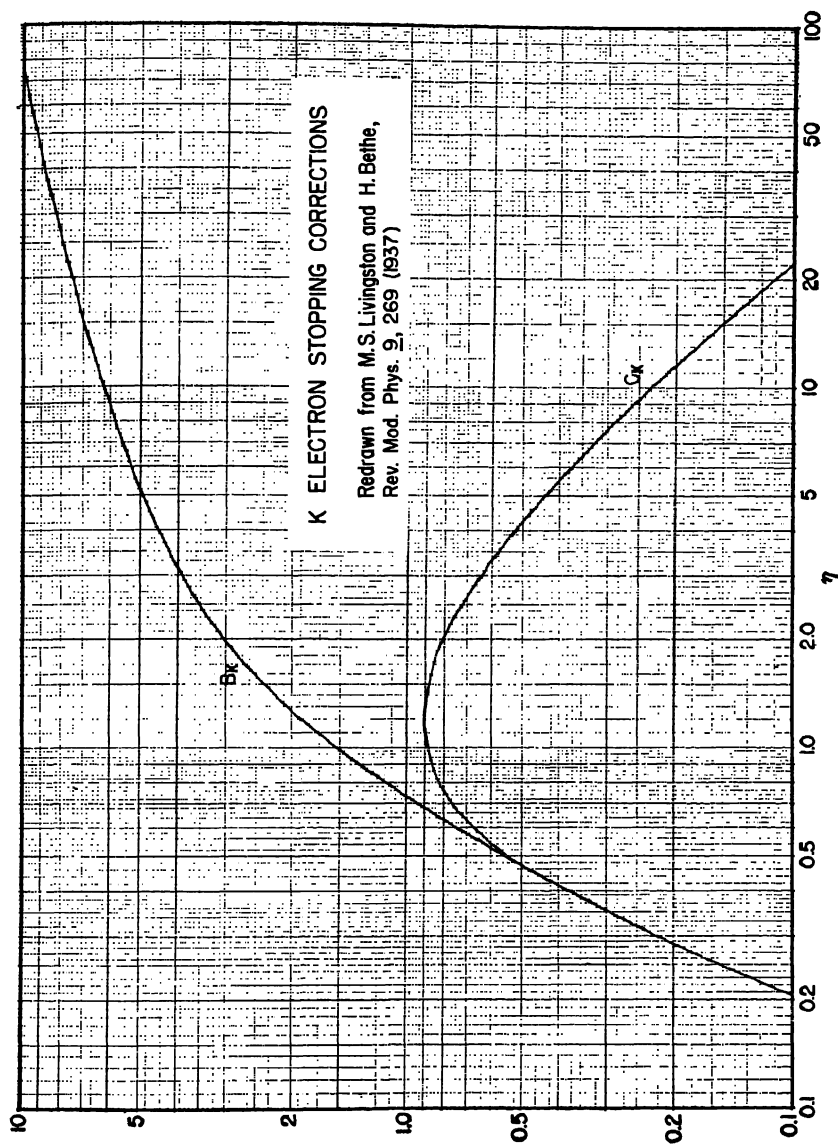


FIG. 15

An approximate value of the effective nuclear charge at the K orbit may be obtained by using Slater's [28] screening constant of 0.3 for 1s electrons; hence,  $Z_{\text{eff}} = Z - 0.3$  where  $Z$  is the actual atomic number. At very low energies the K electrons, except in very light elements, do not contribute to the stopping and  $B_K \rightarrow 0$ . At high energies where  $\eta \gg 1$ ,  $C_K \rightarrow 0$  and there-

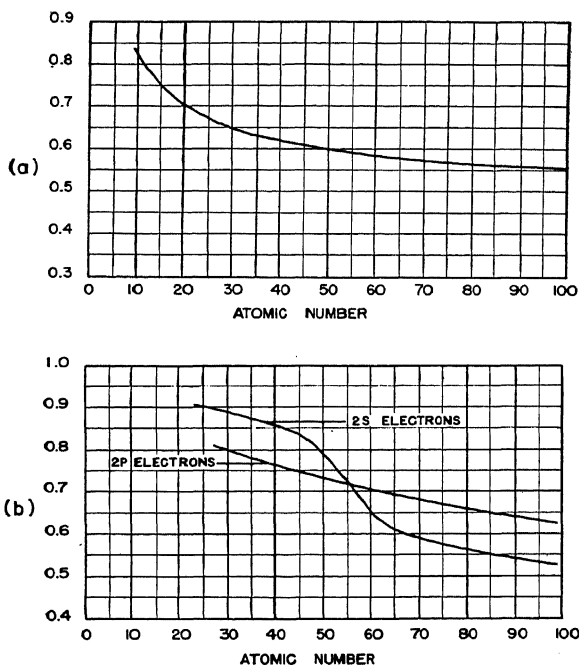


FIG. 16. Effective contribution of each K electron (a), and each L electron (b) to atomic number as a function of  $Z$ . [From H. Hönl, *Z. Phys.*, **84**, 1 (1933).]

fore can be neglected. Consequently at high energies ( $> \sim 10$  mev) the simple stopping formula is valid.

It has also been pointed out by Bethe [6] that in medium and heavy elements all electrons in deep lying shells with ionization potentials greater than  $mE/M$  have little probability for excitation by a passing particle. These electrons (K, L, M, and possibly N) therefore contribute less than one charge unit per electron to the atomic number used in the stopping formula. The contributions, or oscillator strengths, of such electrons have been calculated by Hönl [14] for the K and L electrons and are plotted in Fig. 16. The effective value of  $Z$  in the stopping formula is then  $Z_o = f_K n_K + f_L n_L + \text{remainder of electrons}$ , where  $f$  is the oscillator strength of an electron in the



K or L shell and  $n$  is the number of K or L electrons. This correction is similar in its effect to the procedure followed by Duncanson [9] in treating both  $I$  and  $Z$  as empirical parameters in adjusting the simple stopping formula to the observed data. For very high energies, however, the correction is not important and the actual value of  $Z$  can be used.

**4.4. Stopping Formula for High-energy Particles.** An important correction to the simple stopping formula was introduced by Fermi [15] for particles with very great energy traversing condensed materials. The effect that necessitates the correction arises from changes induced in the field of the passing particle due to local polarization of atoms in media that possess dielectric properties, or more specifically, media with a dielectric constant differing from unity. The rate of energy loss under these conditions is found to be smaller than that computed from the simple relativistic stopping formula which at very high energies increases as the logarithm of energy. The older stopping theories considered the field of the particle to be independent of the properties of the absorbing media. Thus, as a result of the Lorentz contraction of the particle's field, the collision radius, and consequently also the rate of energy loss, increases without limit as  $v \rightarrow c$ . This concept is no longer valid when polarization of the medium is taken into account. When the field of the particle is analyzed into its Fourier components, it is apparent that when  $v > c\epsilon^{-1/2}$  each component of the field is propagated with a different velocity in the medium, or is subject to dispersion. Those frequency components with velocities less than  $v$  form wave fronts or bow waves at various angles with the direction of the particle. As  $v \rightarrow c$  the field is greatly altered as a result of dispersion but, most important, it approaches a limiting form in which all frequency components assume fixed phase relations. The distance at which atoms can be excited by the limiting form of the field remains finite and, correspondingly, the rate of energy loss approaches asymptotically a finite limiting value.

In accordance with this description the reduction in rate of energy loss from the value indicated by the simple stopping formula becomes most important when the velocity of the particle exceeds  $c\epsilon^{-1/2}$ . It is apparent that for electrons the effect of polarization should become appreciable for energies greater than several mev. The correction is of little use in this case, however, because energy loss by radiative collision also becomes significant and a stopping formula based only on ionization energy loss is no longer justified. This applies, fortunately, only to electrons, and the stopping formula can be validly used, so far as is known, for heavier charged particles for all practicable energies. The Fermi corrections, therefore, must be adapted in the appropriate energy range. In general, the corrections become appreciable for protons and alpha particles with  $E \sim 10^4$  mev and for mesons with  $E \sim 10^3$  mev.

The model used by Fermi as the basis for calculating the reduction in energy loss due to polarization is that in which electrons are regarded as classical oscillators and the influence of damping and conduction electrons is negligible. Further, it is assumed that each absorbing substance can be characterized by a single dispersion frequency  $\nu_0$ , which can be obtained from spectroscopic and x-ray data. With these assumptions the dielectric constant of the absorbing medium may be expressed in the simple form

$$\epsilon = 1 + \frac{4\pi ne^2}{m\nu_0^2}$$

When Fermi's corrections are combined with the simple stopping formula, the expressions applicable to particles with very high energies are [15]

For  $v < c\epsilon^{-1/2}$ ,

$$-\frac{dE}{dx} = \frac{2\pi NZe^4z^2}{mv^2} \left[ \log \frac{mv^2W}{I^2} - \log (1 - \beta^2) + (1 - \beta^2) - \log \epsilon \right]$$

For  $v > c\epsilon^{-1/2}$ ,

$$-\frac{dE}{dx} = \frac{2\pi NZe^4z^2}{mv^2} \left[ \log \frac{mv^2W}{I^2} + 1 - \log (\epsilon - 1) - \frac{1 - \beta^2}{\epsilon - 1} \right]$$

where  $I$  = average excitation potential

$\epsilon$  = dielectric constant

The factor  $W$  is the maximum energy that can be transferred to an electron. According to Bhabha [13],  $W$  has the form

$$W = \frac{2m(E^2 - M^2c^4)}{m^2c^2 + M^2c^2 + 2mE}$$

where  $m$  = mass of electron

$M$  = mass of particle

$E$  = energy of particle

$c$  = velocity of light

The correction contained in the first formula is always negligible either when  $v \ll c\epsilon^{-1/2}$  or when the particle passes through media of low density, *i.e.*, with  $\epsilon \simeq 1$ . Under these conditions it reduces to the simple stopping formula.

The second expression diverges rapidly from the simple stopping formula for increasing velocity, and as  $v \rightarrow c$  it approaches asymptotically the constant value given by the expression [15]

$$-\frac{dE}{dx} = \frac{2\pi NZe^4z^2}{mc^2} \left( \log \frac{m^2c^2W}{ZN e^2 h^2} + 1 \right)$$

where  $h$  = Planck's constant

Recently a more general treatment of stopping for charged particles with very high energies was formulated by Halpern and Hall [26]. Their theory, based on a multiple-dispersion-frequency model in which the effects of oscillation damping and conduction electrons are also considered, is a generalization of Fermi's development. The correction in energy loss when these additional factors are taken into account is found to be somewhat less than in the Fermi theory, but the absolute increase in the rate of energy loss is not so fast as  $\log E$  required by the simple stopping formulas.

**4.5. Relative Stopping Power.** Calculations of the stopping power of various substances relative to air are complicated by the dependence on the velocity of the particle as well as on the atomic number and average excitation potential of the absorber. Approximately, the ratio is given by

$$S = \frac{N'Z' \log (2mv^2/I')}{NZ \log (2mv^2/Ie)}$$

where the primed letters refer to the substance and the unprimed to air. Measurements of relative stopping powers of absorbers for alpha particles relative to air are given in Table 10 for two energies and in Fig. 17 for various energies. For heavy charged particles with energies greater than 1 mev the relative stopping power changes very slowly, while above 10 mev it remains essentially constant.

TABLE 10. ATOMIC STOPPING POWER FOR ALPHA PARTICLES RELATIVE TO AIR AT NORMAL TEMPERATURE AND PRESSURE

Absorber	RaC $\alpha$ [16]	6 mev $\alpha$ [10]	Absorber	RaC $\alpha$ [16]	6 mev $\alpha$ [10]
H	0.200	0.20	Ni	1.89	
He	0.308	0.35	Cu	2.00	2.57
Li	0.519	0.50	Zn	2.05	
Be	0.750		Br	2.51	
C	0.814		Kr	....	2.92
N	0.939	0.99	Mo	....	3.20
O	1.000	1.07	Ag	2.74	3.36
Ne	.....	1.23	Cd	2.75	
Mg	1.23		Sn	2.86	3.59
Al	1.27	1.5	I	3.55	
Si	1.23		Ye	....	3.76
Cl	1.76		Pt	3.64	
A	1.80	1.94	Au	3.73	4.50
Ca	1.69		Tl	3.76	
Fe	1.96		Pb	3.86	4.43

**4.6. Atomic Stopping Power.** The atomic stopping power  $\sigma$ , frequently termed stopping cross section, is defined as the energy loss per centimeter

divided by the number of atoms per cubic centimeter. Using an approximate form of the energy-loss formula as an example,  $\sigma$  is given by

$$\sigma = \frac{4\pi e^4 z^2 Z}{mv^2} \log \frac{2mv^2}{1} \quad \text{ev/cm}^2$$

**4.7. Mass Stopping Power.** The mass stopping power is defined as the energy loss per centimeter divided by the density of the absorber or as the stopping power per unit density. From Bragg's empirical rule as well as

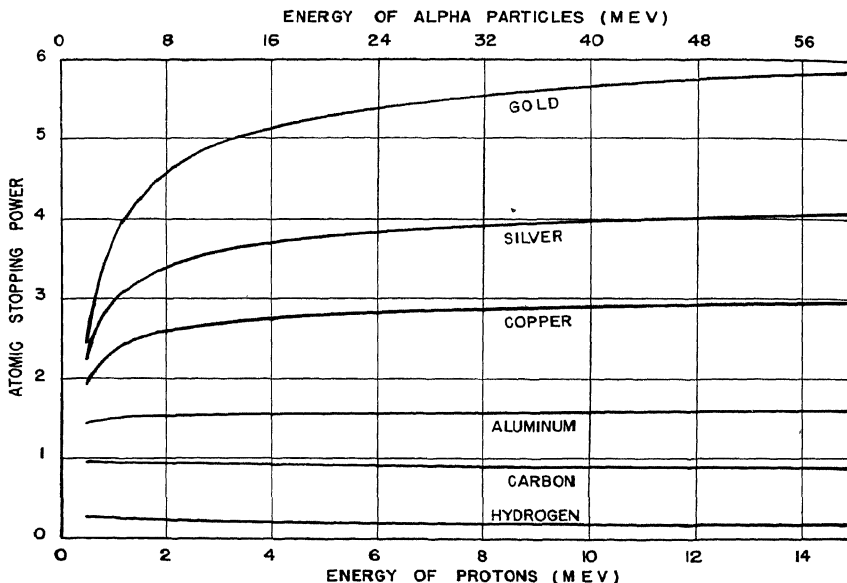


FIG. 17. Atomic stopping power of various elements relative to air.

from the stopping formula, it is found to vary approximately as the inverse square root of the atomic mass of the absorber.

**4.8. Range of Heavy Charged Particles.** The range of a charged particle in an absorber is the total path length traversed before the particle is brought to rest by complete loss of its kinetic energy. With an initial energy  $E_0$ , in units of  $Mc^2$ , the range is calculated with the aid of the energy-loss formula by the integral

$$R = \int_0^{E_0} \frac{dE}{-dE/dx} \quad \text{cm}$$

where  $-dE/dx$  = energy loss per unit length of path

The evaluation of the integral must be carried out numerically, and when possible the integration is usually started at some well-established experimental value of the range and energy rather than that at zero kinetic energy, thus avoiding the error introduced by the poor fit of the stopping formula at very low energies where electron capture and loss become effective.

Unlike the comparatively indeterminate range of beta particles and the exponential absorption of gamma rays, the ranges of heavy charged particles are well defined in that all particles of an initially monoenergetic beam are brought to rest after traversing the same distance through an absorber, with only a small statistical spread or straggling about the mean range. On the other hand, heavier particles with a higher charge number such as fission fragments do not exhibit well-defined ranges. Their charge varies rapidly with velocity due to electron capture, and elastic collisions with nuclei play a more important part; hence, the stopping formula cannot be applied even for a first approximation. It is probable, however, that at very great energies, in the order of 1,000 mev or more depending upon the mass of the particle, the stopping formula would also be applicable to these particles. At such energies the velocity would be great enough to satisfy the conditions required by the formula, and the charge would then remain constant. However, even at such energies it is doubtful that the stopping formula will prove to be very useful since the range of a highly charged particle is small and the straggling large.

At the present time the most accurately measured ranges are those of alpha particles from the natural radioactive isotopes. They have been measured repeatedly by numerous investigators over a period of many years and now stand as the final test of the accuracy of range calculations, at least for energies up to 10 mev.

On the basis of early measurements of the range of alpha particles, three empirical relations were established for the variation of range in air as a function of energy:

1. Range between 0 to 3 cm,

$$R \sim v^{3\frac{1}{2}} \sim E^{\frac{3}{4}} \quad \text{cm}$$

2. Range between 3 to 7 cm (Geiger formula),

$$R = 9.67 \times 10^{-28} v^3 \quad \text{cm}$$

3. Range above 7 cm,

$$R \sim v^4 \sim E^2 \quad \text{cm}$$

These relations should be regarded only as qualitative, because in the variation of  $R$  with  $v^n$ , the exponent  $n$  also depends on the velocity and increases from  $n = 1.4$  for low velocities to  $n = 4.0$  for very high velocities [6].

The relative range of a particle in a substance compared with its range in

air cannot be calculated with accuracy by a simple expression because of the involved dependence on the stopping power of different atoms. Nevertheless, when the range in air is known, a rough estimate of the range of alpha particles in substances other than air can be calculated from Geiger's formula in the energy interval for which it is valid.

$$R = 3.2 \times 10^{-4} \frac{R_o \sqrt{A}}{\rho} \quad \text{cm}$$

where  $R_o$  = range in air in cm

$\rho$  = density of absorber

$A$  = average atomic weight of absorber

Proton ranges have not been investigated so exhaustively as those of alpha particles but can be computed from the range formula with somewhat greater accuracy at lower energies because of the smaller straggling; the minimum velocity conditions for protons holds for energies as low as  $\sim 0.2$  mev. However, if the alpha-particle range in a substance is known, the range of a proton with the same initial velocity can be obtained from the relation

$$R_p = \frac{M_p}{M_\alpha} \left( \frac{z_\alpha}{z_p} \right)^2 R_\alpha + \text{const} \quad \text{cm}$$

TABLE 11. RANGES OF RaC' ALPHA PARTICLES IN VARIOUS SUBSTANCES [16]

Substance	Range $\times 10^{-3}$ cm	Substance	Range $\times 10^{-3}$ cm
Li	12.91	Ag	1.92
Mg	5.78	Cd	2.42
Al	4.06	Sn	2.94
Ca	7.88	Pt	1.28
Fe	1.87	Au	1.40
Ni	1.84	Tl	2.33
Cu	1.83	Pb	2.41
Zn	2.28		

where  $M_p$ ,  $z_p$ ,  $M_\alpha$ ,  $z_\alpha$  = mass and charge of proton and alpha particle, respectively

The constant is included to adjust the range at very low energies since the straggling of alpha particles is greater than that of protons. Blackett and Lees [23] have evaluated the constants and give the range relation as

$$R_p = 1.0072 R_\alpha - 0.20 \quad \text{cm}$$

In a similar way deuteron ranges can be obtained from the ranges of alpha particles with the same initial velocity from the relation

$$R_d = \frac{M_d}{M_\alpha} \left( \frac{z_d}{z_\alpha} \right)^2 R_\alpha + \text{const}$$

$$R_d = 2.012 R_\alpha \quad \text{cm}$$

**4.9. Specific Ionization.** The number of ion pairs formed by a charged particle per unit length of path is referred to as the specific ionization. Its value depends upon the absorbing medium and on the velocity and charge of the particle at any instant. The most intense ionization, as may be seen from the rate of energy loss indicated by the stopping formula, is produced near the end of the particle's range where the velocity is low. A maximum

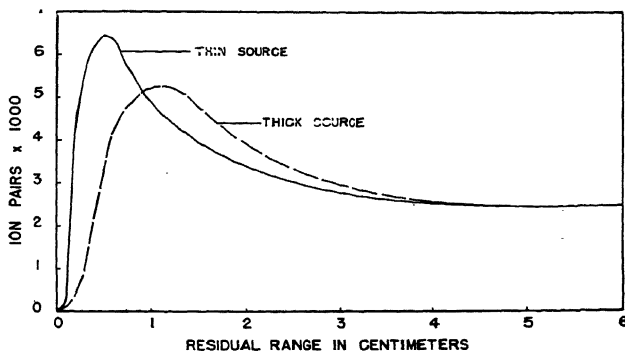


FIG. 18. Bragg curve of specific ionization by alpha particles in air.

value is reached and then drops rapidly to zero as the particle is brought to rest. At high energies the specific ionization remains relatively constant at one-third to one-tenth of its maximum value near the end of the particle's range. In air at normal temperature and pressure the specific ionization of alpha particles, for example, remains essentially constant with a value of roughly 2,000 ion pairs per centimeter for high energies. At low energies, it increases rapidly to a maximum of  $\sim 6,000$  ion pairs for an energy corresponding to a residual range of approximately 0.4 cm, as shown in Fig. 18. Similar curves hold for protons and deuterons.

The average energy  $W$  spent by charged particles in forming one ion pair has been subjected to extensive experimental and theoretical investigation. It is found that for energies greater than about 2.0 mev the value of  $W$  for protons and alpha particles remains essentially constant although there is a very weak dependence on velocity. In an exhaustive survey of the results obtained by various investigators, Gray [29] proposed the value of  $W = 36$  ev per ion pair formed in air by protons and  $W = 31.5$  ev per ion pair formed by alpha particles (measured with alpha particles from RaC'). For hydrogen

the values of  $W$  for protons and alpha particles are  $35 \pm 1.5$  ev per ion and 36 ev per ion, respectively (see also Table 12).

TABLE 12. AVERAGE IONIZATION ENERGY REQUIRED TO FORM ONE ION PAIR BY ALPHA PARTICLES (7 mev) [31]

Absorber.....	N <sub>2</sub>	O <sub>2</sub>	Air	Cl <sub>2</sub>	CO <sub>2</sub>	BF <sub>3</sub>	CH	CF <sub>4</sub>
$W$ , ev.....	33.7	28.5	32.1	25.0	31.2	33.3	27.4	31.1

**4.10. Delta Rays.** The recoil electrons produced by alpha particles, observed in a Wilson cloud chamber as short, faint straggling tracks branching from the alpha track, are referred to as delta rays after J. J. Thomson [17]. The maximum energy imparted to a recoil electron, which occurs in a head-on collision, corresponds to twice the alpha-particle velocity or

$$E_{\max} = 2mv^2 = \frac{4mE}{M}$$

Thus, for a 10-mev alpha particle the most energetic delta ray will have an energy of approximately 6,300 ev and a range in air of several millimeters. The average number of ion pairs liberated by a delta particle of energy  $E_d$  has been calculated to be [18]

$$n = \frac{3}{4} \left( 1 + \frac{E_d}{I} \right)$$

where  $I$  = ionization potential of absorber

The number of delta rays produced and the distribution of their energy depends in a complicated way on the particle velocity and on the kind of absorber, and no satisfactory method for these calculations is available.

**4.11. Straggling of Charged Particles.** The observed ranges of initially monoenergetic particles of the same mass and charge exhibit a statistical fluctuation or "straggling" about an average value. Two factors are mainly responsible for straggling: (1) fluctuation in the number of ions produced per unit path length, particularly near the end of the range where the charge of the particle fluctuates; (2) statistical variation in the energy loss per ion pair. By plotting the number of particles in a beam against distance from the source, an integral range distribution or particle-range curve is obtained, as shown in Fig. 19. Differentiating this curve results in a differential number-range distribution curve which is the distribution of ranges about an average value  $R_0$ . Because of the statistical nature of straggling this curve follows to a close approximation a Gaussian distribution given by the following



expression, provided that the beam of particles was monoenergetic initially [27]:

$$P dR = \frac{1}{\pi^{1/2}\alpha} e^{-(R_0-R)^2/\alpha^2} dR$$

where  $P$  = probability that a particle has a range lying in the interval  $R$  to  $R + dR$

$\alpha$  = range straggling parameter

$R_0$  = mean range

The parameter  $\alpha$  is the half-width of the differential distribution curve at  $1/e$  of its maximum height. It increases with energy, and its order of magnitude may be illustrated by the observed value of approximately  $\alpha = 0.13$  cm for 10-mev alpha particles stopped in air.

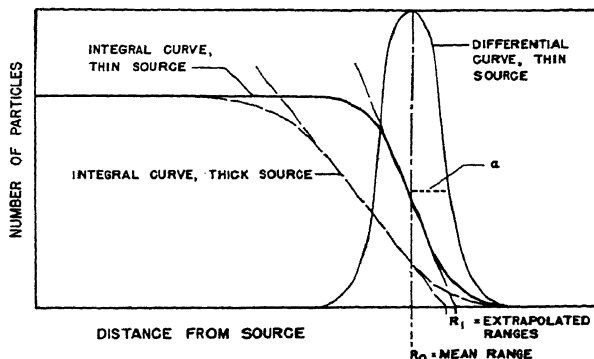


FIG. 19. Integral and differential number-range curves for heavy charged particles.

The mean range  $R_0$  is the distance from the source to the maximum of the differential number-range (Gaussian) curve, or alternatively it may be defined as the distance at which the initial number of particles is reduced to one-half. In practice the extrapolated number-distance range is the most readily determined quantity. This is the range indicated by the intercept of a line extended along the straight portion of the slope of steepest descent of the integral range curve. The mean and the extrapolated ranges are then related by the expression

$$R_{ex} - R_0 = \frac{\pi^{1/2}\alpha}{2}$$

These definitions are exact only for strictly monoenergetic particles such as those emitted from a source that is infinitesimal in thickness. In practice, sources and targets are often "thick" in that their thickness is comparable to or greater than the range of the particles. In this case the beam is not

strictly homogeneous because particles emitted by atoms at various depths are partially absorbed and scattered in different amounts before reaching the surface of the source. However, from thick, homogeneous samples and from thick targets in which the production of particles is uniform throughout the target, the extrapolated range obtained from the measured number-range curve gives a mean range that is representative of particles coming from the surface [6]. Corrections must be applied in all other cases such as "semi-thick" targets and sources where the thickness is comparable to the particle's range and thick targets when the radiation producing the particles does not penetrate the target with uniform intensity.

**4.12. Scattering of Charged Particles.** Elastic scattering of heavy charged particles is quantitatively accounted for on the basis of interaction between the coulomb fields of the incident particle of charge  $ze$  and the struck nucleus of charge  $Ze$ . It is assumed in such collisions that the particle does not approach the nucleus so closely as to be affected by the short-range nuclear forces since these fields result in a different kind of scattering. For alpha particles the minimum collision distance appears to be approximately  $2.05 \times 10^{-13} A^{1/2}$ , where  $A$  is the atomic number of the struck nucleus.

The simplest scattering occurs in collisions of particles with very heavy nuclei, for then the struck nucleus remains virtually stationary during the collision and the particle is deflected with little loss of energy and momentum. Rutherford's formula expressing the number of particles deflected into a unit solid angle at an angle  $\theta$  with the initial direction is then

$$n(\theta) = n_o N \left( \frac{e^2 z Z}{2 m v^2} \right)^2 \text{cosec}^4 \frac{\theta}{2}$$

where  $n_o$  = initial number of particles per  $\text{cm}^2$  of the beam

$N$  = number of atoms per cc

$z$  = charge number of particle

$Z$  = atomic number of scatterer

$m$  = mass of particle

Collisions of particles with nuclei of comparable or smaller mass must be corrected for the contribution of energy to the struck nucleus which recoils with an appreciable fraction of the initial kinetic energy of the incident particle. The general Rutherford formula for the number of particles,  $n$ , scattered into a unit solid angle at an angle  $\theta$  is

$$n(\theta) = n_o N \left( \frac{e^2 z Z}{m v^2} \right)^2 \text{cosec}^3 \theta \frac{[\cot \theta \pm \sqrt{\text{cosec}^2 \theta - (m/M)^2}]^2}{\sqrt{\text{cosec}^2 \theta - (m/M)^2}}$$

where  $n_o$  = initial number of particles per  $\text{cm}^2$  of beam

$M$  = mass of nucleus

$m$  = mass of particle

The + sign is used if  $M > m$ , and - sign if  $M < m$ . This expression obviously reduces to the preceding formula when  $M \gg m$ .

Although particles will be scattered in all directions, the intensity drops off very rapidly with increasing angle as measured from the forward direction, thus in the case of alpha particles on platinum, less than 1 particle in 8,000 will be scattered into an angle greater than 90 deg. Backscattering in light elements, consequently, is entirely negligible. In general, calculations of scattering of particles by heavy elements for energies up to about 10 mev are given to a high degree of accuracy by the Rutherford formulas.

Deviations from Rutherford (coulomb field) scattering calculated from the formulas above will be found for certain energies of the incident particle depending upon the properties of the scattering nucleus. Anomalous scattering will occur if the particle possesses sufficient energy to penetrate the electrostatic potential barrier and be affected by the nuclear forces or if the energy of the particle plus the internal energy of the scattering nucleus equals the energy of a quantum state of the compound nucleus. Anomalous scattering that increases slowly with increased particle energy can usually be identified with penetration of the potential barrier. Anomalous scattering that increases rapidly to a maximum intensity and then decreases as the energy is increased indicates resonance scattering.

**4.13. Alpha Decay.** The theory of alpha decay is based on the now familiar quantum mechanical problem of the penetration of charged particles through potential barriers. Some assumptions must be made about the shape of the potential field forming the barrier, but the theory can account for the main details of alpha emission and provide estimates of its probability.

If it is assumed that the alpha particle already exists as a separate entity within the nucleus and that the only effect of the remaining particles on it is to provide a potential "well" in which the alpha particle exists in some energy state, the problem immediately reduces to the one-body model for alpha decay [19,20].

Within the nucleus it is assumed that alpha particles can move with relative freedom, but at the surface it is constrained from leaving by an effective surface tension maintained by the unsaturated, attractive nuclear forces of those particles lying at the surface. This force rises rapidly to a maximum value at a distance approximately equal to the nuclear radius. Beyond the radius, the nuclear potential field rapidly vanishes, and only the coulomb field due to the protons remains. The height of the potential barrier above the zero coulomb energy, *i.e.*, the value approached asymptotically by the coulomb field at great distances, is approximately the magnitude of the coulomb field at the surface or

$$E_b = \frac{e^2 z Z}{R}$$

where  $z$  = alpha-particle charge

$Z$  = atomic number

$R$  = nuclear radius

The field in the nucleus therefore may be represented by a cylindrical potential well, as shown in Fig. 20, extending an unknown distance  $E_o$  below the zero coulomb energy and a distance  $E_b$  above. Outside the potential well only the coulomb field is effective.

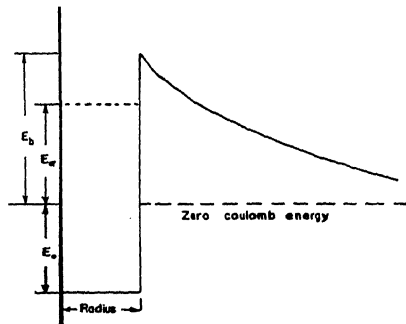


FIG. 20. Diagram of potential fields of nucleus showing "square" potential well and coulomb barrier.

probability of emission per unit time by penetration through the barrier is greatly reduced. The mean life  $\tau$  for alpha emission under this condition (alpha decay) is given by the expression

$$\tau = \tau_o e^f$$

where  $\tau_o \approx 10^{-21}$  sec and the function  $f$  depends on the charge and mass of the nucleus. If the observed kinetic energy of the alpha particle is small compared to  $2e^2Z/R$ , the function  $f$  is given approximately by

$$f = \frac{8\pi^2 e^2 Z}{h} - \frac{8\pi e (4ZmR)^{1/2}}{h}$$

where  $E_a$  = energy of alpha particle above zero coulomb energy, or the observed kinetic energy of the alpha particle plus the recoil nucleus (Fig. 20)

$m$  = mass of alpha particle

$R$  = nuclear radius,  $\approx 1.47 \times 10^{-13} A^{1/2}$  cm

$v$  = alpha-particle velocity

$h$  = Planck's constant

The factor  $\tau_o$  is the mean life for emission in the absence of a barrier and may be thought of as the period of vibration of the alpha particle in the nucleus. The reciprocal of  $e^f$  is the transmission coefficient, or penetrability, and

is the factor that determines the probability of emission through the barrier. It is a sensitive function of the barrier height and width as is readily apparent from the observed half-lives of the natural radioactive isotopes which range from  $10^{-7}$  sec to  $10^{10}$  years.

Writing the expression for  $\tau$  in terms of logarithms,

$$\log \tau = \frac{A}{v} + B$$

where  $A = f\bar{v}$

$B = \log \tau_0$

If it is assumed that the nuclear radius is nearly constant for all alpha emitters,  $A$  and  $B$  are then also nearly constant and the logarithm of the mean life is inversely proportional to the velocity of the alpha particle. A similar relation is found between the range  $R$  and the mean life.

$$\log \tau = C - D \log R$$

or

$$\log \lambda = E \log R + F$$

where  $\lambda$  = disintegration constant

$C, D, E, F$  = constants

These important relations were first discovered empirically by Geiger and Nuttall [21] when they plotted the measured values of  $\lambda$  and  $R$  for the alpha emitters of the natural radioactive series. The components of each series are found to fall remarkably close to straight lines as required by the expression above.

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TABLE 13. RANGE AND ENERGY LOSS OF PROTONS IN HYDROGEN AND HELIUM

The following data were computed for hydrogen and helium at 0°C and 760 mm of mercury from the Bethe stopping theory. Corrections for noncontributing shells, the Fermi effect, and *Bremsstrahlung* are not included. Data calculated by W. Aron and B. G. Hoffman, Radiation Laboratory, University of California, for the Atomic Energy Commission.

Energy, mev	Hydrogen ( $I = 17.5$ ev)		Helium ( $I = 44$ ev)	
	Energy loss, mev/cm	Range, cm	Energy loss, mev/cm	Range, cm
0	0	0	0	0
1	$5.88476 \times 10^{-2}$	$1.0025 \times 10^1$	$4.75985 \times 10^{-2}$	$1.3030 \times 10^1$
2	3.36971		2.80637	
3	2.41487	6.9061	2.00607	8.4160
4	1.90174		1.61918	
5	1.57824	$1.7410 \times 10^2$	1.35183	$2.0778 \times 10^2$
6	1.35435	2.4270	1.16537	2.8769
7	1.18957	3.2169	1.02733	3.7930
8	1.06288	4.1076	$9.20701 \times 10^{-3}$	4.8229
9	$9.62240 \times 10^{-3}$	5.0979	8.35660	5.9645
10	8.80238	6.1855	7.66136	7.2155
11	8.12051		7.08158	
12	7.54400	8.6480	6.59015	$1.0039 \times 10^3$
13	7.04979		6.16793	
14	6.62113	$1.1485 \times 10^3$	5.80097	1.3281
15	6.24557		5.47889	
16	5.91365	1.4687	5.19375	1.6931
17	5.61804		4.93944	
18	5.35301	1.8246	4.71110	2.0980
19	5.11395		4.50487	
20	4.89716	2.2157	4.31764	2.5419
25	4.05773		3.59050	
30	3.48225	4.6776	3.08987	5.3241
35	3.06191		2.72299	
40	2.74078	7.9416	2.44195	8.9943
45	$2.48706 \times 10^{-3}$		$2.21941 \times 10^{-3}$	
50	2.28132	$1.1961 \times 10^4$	2.03861	$1.3498 \times 10^4$
55	2.11097		1.88867	
60	1.96757	1.6697	1.76226	1.8791
65	1.84498		1.65406	
70	1.73905	2.2115	1.56046	2.4736
75	1.64654		1.47862	
80	1.56505	2.8188	1.40644	3.1597
85	1.49262		1.34229	
90	1.42789	3.4886	1.28488	3.9046
95	1.36965		1.23320	
100	1.31697	4.2186	1.18642	4.7154

TABLE 13. RANGE AND ENERGY LOSS OF PROTONS IN HYDROGEN AND HELIUM—(Continued)

Energy, mev	Hydrogen ( $I = 17.5$ ev)		Helium ( $I = 44$ ev)	
	Energy loss, mev/cm	Range, cm	Energy loss, mev/cm	Range, cm
125	1.11433		1.00617	
150	$9.76921 \times 10^{-4}$	9.0290	$8.83659 \times 10^{-4}$	9.6737
175	8.77559		7.94951	
200	8.02397	$1.4386 \times 10^5$	7.27717	$1.5955 \times 10^5$
225	7.43533		6.75029	
250	6.96260	2.1104	6.32684	2.3356
275	6.57481		5.97928	
300	6.25129	2.8704	5.68919	3.1712
325	5.97754		5.44364	
350	5.74314	3.7065	5.23335	4.0893
375	5.54040		5.05144	
400	5.36349	4.6086	4.89271	5.0787
425	5.20796		4.75315	
450	5.02865	5.5697	4.62963	6.1303
475	$4.94771 \times 10^{-4}$		$4.51967 \times 10^{-4}$	
500	4.83799	$6.5814 \times 10^5$	4.42128	$7.2363 \times 10^5$
550	4.65020		4.25293	
600	4.49589	8.7302	4.11470	9.5859
650	4.36739		3.99967	
700	4.25914	$1.1022 \times 10^6$	3.90293	$1.2085 \times 10^6$
750	4.16719		3.82084	
800	4.08836	1.3418	3.75057	1.4701
850	4.02034		3.69004	
900	3.96131	1.5904	3.63760	1.7410
950	3.90981		3.59196	
1,000	3.86469	1.8461	3.55206	2.0194
1,500	3.61964		3.33866	
2,000	3.54362	4.5895	3.27707	4.9940
2,500	3.52729		3.26866	
3,000	3.53620	7.4212	3.28242	8.0499
3,500	3.55670		3.30613	
4,000	3.58263	$1.0232 \times 10^7$	3.33429	$1.1074 \times 10^7$
4,500	3.61094		3.36423	
5,000	3.64006	1.3001	3.39457	1.4046
5,500	3.68530		3.42457	
6,000	3.69769	1.5719	3.45387	1.6967
6,500	3.72551		3.48228	
7,000	3.75246	1.8403	3.50971	1.9839
7,500	3.77849		3.53614	
8,000	$3.80360 \times 10^{-4}$	$2.1050 \times 10^7$	$3.56157 \times 10^{-4}$	$2.2666 \times 10^7$
8,500	3.82779		3.58604	
9,000	3.85111	2.3663	3.60960	2.5456
9,500	3.87358		3.63227	
10,000	3.89525	2.6244	3.65412	2.8209



TABLE 14. RANGE AND ENERGY LOSS OF PROTONS IN LITHIUM AND BERYLLIUM

The following data were computed for lithium and beryllium from the Bethe stopping theory. Corrections for noncontributing shells, the Fermi effect and *Bremsstrahlung* are not included. Data calculated by W. Aron and B. G. Hoffman, Radiation Laboratory, University of California, for the Atomic Energy Commission.

Energy, mev	Lithium ( $I = 34.5$ ev)		Beryllium ( $I = 46$ ev)	
	Energy loss, mev/(mg/cm <sup>2</sup> )	Range, mg/cm <sup>2</sup>	Energy loss, mev/(mg/cm <sup>2</sup> )	Range, mg/cm <sup>2</sup>
0	0	0	0	0
1	$2.58707 \times 10^{-1}$	2.8738	$2.46973 \times 10^{-1}$	2.9100
2	1.51183	8.1422	1.45866	8.4276
3	1.09382	1.60295 $\times 10$	1.06050	1.65443 $\times 10$
4	$8.66545 \times 10^{-2}$	2.63834	$8.42671 \times 10^{-2}$	2.71890
5	7.22183	3.90797	7.03775	4.02471
6	6.21736	5.40540	6.06860	5.56018
7	5.47503	7.12294	5.35088	7.31884
8	4.90241	9.05655	4.79629	9.29626
9	4.44626	1.12012 $\times 10^2$	4.35391	1.14873 $\times 10^2$
10	4.07372	1.35535	3.99217	1.38884
11	3.76331	1.61096	3.69045	1.64959
12	3.50041	1.88670	3.43468	1.93067
13	3.27435	2.18225	3.21490	2.23179
14	3.07860	2.49751	3.02387	2.55269
15	2.90660	2.83183	2.85618	2.89312
16	2.75440	3.18540	2.70771	3.25285
17	2.61871	3.55788	2.57528	3.63168
18	2.49694	3.94910	2.45636	4.02940
19	2.38700	4.35880	2.34896	4.44583
20	2.28721	4.78701	2.25143	4.88079
25	1.90004		1.87264	
30	1.63380	1.00445 $\times 10^3$	1.61178	1.02152 $\times 10^3$
35	1.43889		1.42058	
40	1.28969	1.69902	1.27409	1.72503
45	$1.17162 \times 10^{-2}$		$1.15808 \times 10^{-2}$	
50	1.07576	2.55219 $\times 10^3$	1.06382	2.58817 $\times 10^3$
55	$9.96287 \times 10^{-3}$		$9.85637 \times 10^{-3}$	
60	9.29321	3.55561	9.19723	3.60244
65	8.72019		8.63296	
70	8.22467	4.70211	8.14481	4.76052
75	7.79157		7.71801	
80	7.40969	5.98531	7.34156	6.05595
85	7.07034		7.00694	
90	6.76674	7.39944	6.70749	7.48288
95	6.49347		6.43788	
100	6.24616	8.93925	6.19383	9.03599

TABLE 14. RANGE AND ENERGY LOSS OF PROTONS IN LITHIUM AND BERYLLIUM—(Continued)

Energy, mev	Lithium ( $I = 34.5$ ev)		Beryllium ( $I = 46$ ev)	
	Energy loss, mev/(mg/cm <sup>2</sup> )	Range, mg/cm <sup>2</sup>	Energy loss, mev/(mg/cm <sup>2</sup> )	Range, mg/cm <sup>2</sup>
125	5.29376		5.25348	
150	4.64686	$1.83635 \times 10^4$	4.61426	$1.85324 \times 10^4$
175	4.17853		4.15124	
200	3.82395	3.03134	3.80050	3.05609
225	3.54605		3.52554	
250	3.32275	4.44008	3.30454	4.47303
275	3.13950		3.12314	
300	2.98657	6.03164	2.97173	6.07293
325	2.85714		2.84357	
350	2.74630	7.78077	2.73381	7.83041
375	2.65042		2.63886	
400	2.56675	9.66654	2.55601	9.72443
425	2.49320		2.48317	
450	2.42807	$1.16714 \times 10^5$	2.41870	$1.17374 \times 10^5$
475	$2.37013 \times 10^{-3}$		$2.36131 \times 10^{-3}$	
500	2.31826	$1.37804 \times 10^5$	2.30995	$1.38542 \times 10^5$
550	2.22951		2.22209	
600	2.15662	1.82624	2.14995	1.83512
650	2.09596		2.08993	
700	2.04490	2.30310	2.03943	2.31336
750	2.00157		1.99659	
800	1.96446	2.80251	1.95993	2.81402
850	1.93249		1.92835	
900	1.90476	3.31983	1.90099	3.33245
950	1.88062		1.87719	
1,000	1.85950	3.85146	1.85638	3.86504
1,500	1.74605		1.74518	
2,000	1.71259	9.53948	1.71322	9.55613
2,500	1.70722		1.70900	
3,000	1.71361	$1.53906 \times 10^6$	1.71635	$1.54013 \times 10^6$
3,500	1.72531		1.72887	
4,000	1.73941	2.11859	1.74371	2.11847
4,500	1.75451		1.75947	
5,000	1.76986	2.68852	1.77542	2.68683
5,500	1.78645		1.79119	
6,000	1.79998	3.24846	1.80659	3.24515
6,500	1.81444		1.82151	
7,000	1.82840	3.79956	1.83592	3.79418
7,500	$1.84187 \times 10^{-3}$		$1.84980 \times 10^{-3}$	
8,000	1.85484	$4.34252 \times 10^6$	1.86316	$4.33482 \times 10^6$
8,500	1.86732		1.87601	
9,000	1.87934	4.87808	1.88837	4.86790
9,500	1.89091		1.90028	
10,000	1.90265	5.40695	1.91175	5.39416

TABLE 15. RANGE AND ENERGY LOSS OF PROTONS IN BORON AND CARBON

The following data were computed for boron and carbon from the Bethe stopping theory. Corrections for noncontributing shells, the Fermi effect and *Bremsstrahlung* are not included. Data calculated by W. Aron and B. G. Hoffman, Radiation Laboratory, University of California, for the Atomic Energy Commission.

Energy, mev	Boron ( $I = 57.5$ ev)		Carbon ( $I = 69$ ev)	
	Energy loss, mev/(mg/cm <sup>2</sup> )	Range, mg/cm <sup>2</sup>	Energy loss, mev/(mg/cm <sup>2</sup> )	Range, mg/cm <sup>2</sup>
0	0	0	0	0
1	$2.42467 \times 10^{-1}$	2.890	$2.41758 \times 10^{-1}$	2.760
2	1.44542	8.4650	1.49676	8.1130
3	1.05529	$1.66485 \times 10$	1.09761	$1.59838 \times 10$
4	$8.40700 \times 10^{-2}$	2.73305	$8.75831 \times 10^{-2}$	2.62925
5	7.03397	4.04064	7.33961	3.88859
6	6.07363	5.57469	6.34518	5.35789
7	5.36113	7.33162	5.60616	7.03867
8	4.80635	9.30499	5.03354	8.92378
9	4.36942	$1.14905 \times 10^2$	4.57573	$1.10106 \times 10^2$
10	4.00899	1.38826	4.20066	1.32935
11	$3.70811 \times 10^{-2}$	1.64782	3.88732	
12	3.45285	1.92749	3.62130	1.84374
13	3.23335	2.22697	3.39243	
14	3.04245	2.54597	3.19326	2.43321
15	2.87479	2.88425	3.01826	
16	2.72627	3.24160	2.86317	3.09577
17	2.59374	3.61779	2.72471	
18	2.47468	4.01262	2.60039	3.82973
19	2.36710	4.42591	2.48783	
20	2.26939	4.85748	2.38565	4.63358
25	1.88953		1.98812	
30	1.62761	$1.01441 \times 10^3$	1.71372	9.65798
35	1.43545		1.51222	
40	$1.28811 \times 10^{-2}$	$1.71063 \times 10^3$	$1.35762 \times 10^{-2}$	$1.62667 \times 10^3$
45	1.17135		1.23504	
50	1.07642	2.56400	1.13533	2.43603
55	$9.97656 \times 10^{-3}$		1.05256	
60	9.31220	3.56604	$9.82726 \times 10^{-3}$	3.38580
65	8.74326		9.22900	
70	8.25089	4.70951	8.71124	4.46908
75	7.82082		8.25809	
80	7.44035	5.98793	7.85829	5.67979
85	7.10259		7.50276	
90	6.80024	7.39565	7.18446	7.01242
95	6.52797		6.89777	
100	6.28146	8.92731	6.63817	8.46197

TABLE 15. RANGE AND ENERGY LOSS OF PROTONS IN BORON AND CARBON—(Continued)

Energy, mev	Boron ( $I = 57.5$ ev)		Carbon ( $I = 69$ ev)	
	Energy loss, mev/(mg/cm <sup>2</sup> )	Range, mg/cm <sup>2</sup>	Energy loss, mev/(mg/cm <sup>2</sup> )	Range, mg/cm <sup>2</sup>
125	5.33120		5.63703	
150	4.68484	$1.82853 \times 10^4$	4.95568	$1.73119 \times 10^4$
175	4.21641		4.46169	
200	3.86146	3.01278	4.08726	2.85450
225	3.58311		3.79356	
250	3.35934	4.40695	3.55741	4.16715
275	3.17563		3.35463	
300	3.02229	5.98040	3.20164	5.65534
325	2.89247		3.06459	
350	2.78129	7.70817	2.94721	7.28607
375	2.68510		2.85566	
400	2.60117	9.56958	2.75705	9.03835
425	2.52738		2.67915	
450	$2.46208 \times 10^{-3}$	$1.15473 \times 10^5$	$2.61020 \times 10^{-3}$	$1.09040 \times 10^5$
475	2.40394		2.54883	
500	2.35192	1.36267	2.49391	1.28652
550	2.26294		2.39999	
600	2.18989	1.80424	2.32290	1.70288
650	2.12914		2.25880	
700	2.07803	2.2737	2.20489	2.14536
750	2.03470		2.15920	
800	1.99763	2.76500	2.12012	2.60832
850	1.96571		2.08649	
900	1.93808	3.27353	2.05739	3.08746
950	1.91405		2.03209	
1,000	1.89306	3.79587	2.01001	3.57945
1,500	1.781369		1.89296	
2,000	1.74998	9.37126	1.86071	8.82617
2,500	1.74663		1.85803	
3,000	1.75493	$1.50904 \times 10^5$	1.86757	$1.42023 \times 10^5$
3,500	1.76841		1.88251	
4,000	1.78417	2.07442	1.89981	1.95134
4,500	1.80080		1.91798	
5,000	1.81758	2.62974	1.93626	2.47274
5,500	1.83414		1.95427	
6,000	1.85028	3.17499	1.97432	2.98436
6,500	1.86591		1.98878	
7,000	$1.88099 \times 10^{-3}$	$3.71096 \times 10^5$	$2.00513 \times 10^{-3}$	$3.48711 \times 10^5$
7,500	1.89550		2.02087	
8,000	1.90947	4.23856	2.03601	3.98199
8,500	1.92290		2.05041	
9,000	1.93577	4.75864	2.06455	4.46971
9,500	1.94825		2.07802	
10,000	1.96023	5.27195	2.09099	4.95096

TABLE 16. RANGE AND ENERGY LOSS OF PROTONS IN NITROGEN AND OXYGEN

The following data were computed for nitrogen and oxygen at 0°C and 760 mm of mercury from the Bethe stopping theory. Corrections for noncontributing shells, the Fermi effect and *Bremsstrahlung* are not included. Data calculated by W. Aron and B. G. Hoffman, Radiation Laboratory, University of California, for the Atomic Energy Commission.

Energy, mev	Nitrogen ( $I = 80.5$ ev)		Oxygen ( $I = 92$ ev)	
	Energy loss, mev/cm	Range, cm	Energy loss, mev/cm	Range, cm
0		0		0
1	$2.81579 \times 10^{-1}$	2.220	$3.08769 \times 10^{-1}$	1.990
2	1.70599	7.4883	1.88442	
3	1.25422	$1.3877 \times 10$	1.39013	$1.25434 \times 10$
4	1.00377	2.2841	1.11442	2.06241
5	$8.42394 \times 10^{-2}$	3.3775	$9.36501 \times 10^{-2}$	3.04653
6	7.29050	4.6577	8.11301	4.19685
7	6.44692	6.1192	7.17990	5.51021
8	5.79250	7.7584	6.45524	6.98123
9	5.26878	9.5707	5.87478	8.60730
10	4.83937	$1.15531 \times 10^2$	5.39848	$1.03847 \times 10^2$
11	4.48037	1.37025	5.00002	1.23112
12	4.17540	1.60162	4.66135	1.43840
13	3.91288	1.84917	4.36967	1.66012
14	3.68432	2.11268	4.11561	1.89604
15	3.48340	2.39194	3.89218	2.14601
16	3.30528	2.68676	3.69403	2.40982
17	3.14674	2.99697	3.51701	2.68733
18	3.00320	3.32232	3.35784	2.97843
19	2.87391	3.66287	3.21389	3.28295
20	2.75641	4.01819	3.08302	3.60069
25	2.29893		2.57320	
30	1.98284	8.36328	2.22063	7.48263
35	1.75056		1.96136	
40	$1.57223 \times 10^{-2}$	$1.40722 \times 10^3$	$1.76220 \times 10^{-2}$	$1.25780 \times 10^3$
45	1.43076		1.60414	
50	1.31565	2.10586	1.47547	1.88092
55	1.22005		1.36858	
60	1.13936	2.92525	1.27833	2.61139
65	1.07022		1.20098	
70	1.01036	3.85941	1.13399	3.44384
75	$9.57973 \times 10^{-3}$		1.07537	
80	9.11738	4.90309	1.02361	4.37358
85	8.70613		$9.77534 \times 10^{-3}$	
90	8.33789	6.05152	9.36328	5.39637
95	8.00616		8.99176	
100	7.70574	7.30039	8.65525	6.50835
125	6.54676		7.35665	

TABLE 16. RANGE AND ENERGY LOSS OF PROTONS IN NITROGEN AND OXYGEN—(Continued)

Energy, mev	Nitrogen ( $I = 80.5$ ev)		Oxygen ( $I = 92$ ev)	
	Energy loss, mev/cm	Range, cm	Energy loss, mev/cm	Range, cm
150	5.75759	$1.49208 \times 10^4$	6.47202	$1.32898 \times 10^4$
175	5.18522		5.83020	
200	4.75126	2.45506	5.34346	2.18543
225	4.41080		4.96152	
250	4.13700	3.58761	4.65432	3.19226
275	3.91216		4.40202	
300	3.72444	4.86483	4.19136	4.32736
325	3.56551		4.01299	
350	3.42937	6.26646	3.86019	5.57270
375	3.31159		3.72800	
400	3.20881	7.77573	3.61265	6.95639
425	3.11845		3.50910	
450	$3.03849 \times 10^{-3}$	$9.37860 \times 10^4$	$3.42149 \times 10^{-3}$	$8.38053 \times 10^4$
475	2.96731		3.34160	
500	2.90363	$1.10632 \times 10^5$	3.27013	9.87644
550	2.79471		3.14791	
600	2.70533	1.46387	3.04763	$1.30508 \times 10^5$
650	2.63102		2.96427	
700	2.56855	1.84375	2.89420	1.64225
750	2.51561		2.83485	
800	2.47035	2.24112	2.78411	1.99487
850	2.43142		2.74048	
900	2.39774	2.65229	2.70276	2.35970
950	2.36848		2.66999	
1,000	2.34294	3.07441	2.64141	2.73416
1,500	2.20809		2.49096	
2,000	2.17159	7.57273	2.45093	6.72149
2,500	2.16934		2.44927	
3,000	2.18120	$1.21778 \times 10^6$	2.76339	$1.08000 \times 10^6$
3,500	2.19927		2.48441	
4,000	2.22001	1.67243	2.50837	1.48244
4,500	2.24171		2.53335	
5,000	2.26350	2.11853	2.55840	1.87720
5,500	2.28493		2.58300	
6,000	2.30578	2.55621	2.60691	2.26434
6,500	2.32593		2.63001	
7,000	2.34534	2.98618	2.65226	2.64462
7,500	$2.36403 \times 10^{-3}$		$2.67366 \times 10^{-3}$	
8,000	2.38198	$3.40922 \times 10^6$	2.69421	$3.01867 \times 10^6$
8,500	2.39924		2.71325	
9,000	2.41584	3.82604	2.73296	3.38722
9,500	2.43180		2.75123	
10,000	2.44718	4.23728	2.76882	3.75071

TABLE 17. RANGE OF PROTONS IN ALUMINUM

Range data for energies 0.117 to 1.842 mev reported by D. B. Parkinson, R. G. Herb, J. C. Bellamy, and C. M. Hudson, *Phys. Rev.*, **52**, 75 (1937). Range is given in centimeters and distinguished by an asterisk \*. Range data for energies from 1 to 13 mev are taken from M. S. Livingston, and H. Bethe, *Rev. Mod. Phys.*, **9**, 263 (1937). Data for energies 13 to 10,000 mev are calculated from Bethe theory by J. H. Smith, *Phys. Rev.*, **71**, 32 (1947). Bethe correction  $C_K$  is included, but Fermi effect is not taken into account. Average excitation potential,  $I = 150$  ev.

Energy, mev	Energy loss, mev/(mg/cm <sup>2</sup> )	Range, mg/cm <sup>2</sup>	Energy, mev	Energy loss, mev/(mg/cm <sup>2</sup> )	Range, mg/cm <sup>2</sup>
0.117*	.....	$0.979 \times 10^{-4}$	25	1.682	8.369
0.166*	.....	1.44	30	1.456	11.57
0.267*	.....	2.51	35	1.289	15.23
0.342*	.....	3.46	40	1.160	19.33
0.433*	.....	4.67	45	1.058	23.85
0.486*	.....	5.39	50	0.9743	28.78
0.520*	.....	5.93	60	$0.8458 \times 10^{-2}$	$39.83 \times 10^2$
0.630*	.....	7.69	70	0.7516	52.40
0.745*	.....	9.65	80	0.6794	66.42
1.055*	.....	15.6	90	0.6222	81.82
1.393*	.....	23.8	100	0.5757	98.54
1.842*	.....	37.6	120	0.5047	135.8
1	.....	3.45	140	0.4530	177.7
1.5	.....	6.69	160	0.4136	224.0
2	$11.5 \times 10^{-2}$	10.8	180	0.3826	274.3
2.5	9.85	15.6	200	0.3576	328.4
3	8.62	21.0	250	0.3120	478.7
3.5	7.69	27.3	300	0.2813	648.0
4	6.96	34.5	350	0.2593	833.4
4.5	6.37	42.1	400	0.2428	1033
5	5.88	50.3	500	$2.201 \times 10^{-3}$	$1.467 \times 10^4$
5.5	5.47	59.0	600	2.054	1.938
6	5.12	69.1	700	1.952	2.438
6.5	4.82	79.2	800	1.879	2.961
7	4.55	90.0	850	1.851	3.229
7.5	4.31	101.3	900	1.826	3.501
8	4.10	113.2	950	1.802	3.777
8.5	3.92	125.6	1,000	1.785	4.055
9	3.75	138.8	1,250	1.721	5.484
9.5	3.59	152.4	1,500	1.688	6.952
10	3.45	166.7	1,750	1.671	8.441
10.5	3.32	181.4	2,000	1.664	9.941
11	3.21	196.6	2,250	1.663	11.44
11.5	3.10	212.5	2,500	1.665	12.95
12	2.99	229.0	2,750	1.670	14.45
12.5	2.90	246.1	3,000	1.677	15.94
13	2.816	263.7	4,000	1.710	21.85
13.5	2.734	281.8	5,000	1.747	27.63
14	2.659	300.6	6,000	1.782	33.30
15	2.518	$3.393 \times 10^2$	7,000	1.815	38.86
17	2.281	4.228	8,000	1.845	44.32
19	2.089	5.146	9,000	1.873	49.70
21	1.930	6.143	10,000	1.898	55.01
23	1.796	7.218			

TABLE 18. RANGE OF PROTONS IN PARAFFIN  $(CH_2)_n$ 

Range-energy data calculated from the Bethe theory by J. O. Hirschfelder, and J. L. Magee, *Phys. Rev.*, **73**, 207 (1948).

Energy, mev	Range, mg/cm <sup>2</sup>	Energy, mev	Range, mg/cm <sup>2</sup>
0.005	0.02322	1.8	5.23363
0.010	0.03197	1.9	5.74061
0.015	0.03732	2.0	6.26829
0.02	0.04143	2.1	6.81661
0.03	0.04824	2.2	7.38533
0.04	0.05462	2.3	7.97427
0.05	0.06096	2.4	8.58326
0.10	0.09613	2.5	9.21214
0.15	0.13860	2.6	9.86087
0.20	0.18908	2.7	10.52932
0.3	0.31370	2.8	11.21735
0.4	0.46833	2.9	11.92483
0.5	0.65115	3.0	12.65163
0.6	0.86101	3.5	16.57086
0.7	1.09677	4.0	20.95967
0.8	1.35759	4.5	25.81139
0.9	1.64291	5.0	31.11793
1.0	1.95202	5.5	36.87259
1.1	2.28446	6.0	43.06963
1.2	2.63976	6.5	49.70417
1.3	3.01756	7.0	56.77056
1.4	3.41751	7.5	64.26439
1.5	3.83930	8.0	72.18204
1.6	4.28273	8.5	80.52013
1.7	4.74757	9.0	89.27502
		10.0	108.0222
		11.0	128.4035
		12.0	150.4002
		13.0	173.9955
		14.0	199.1717



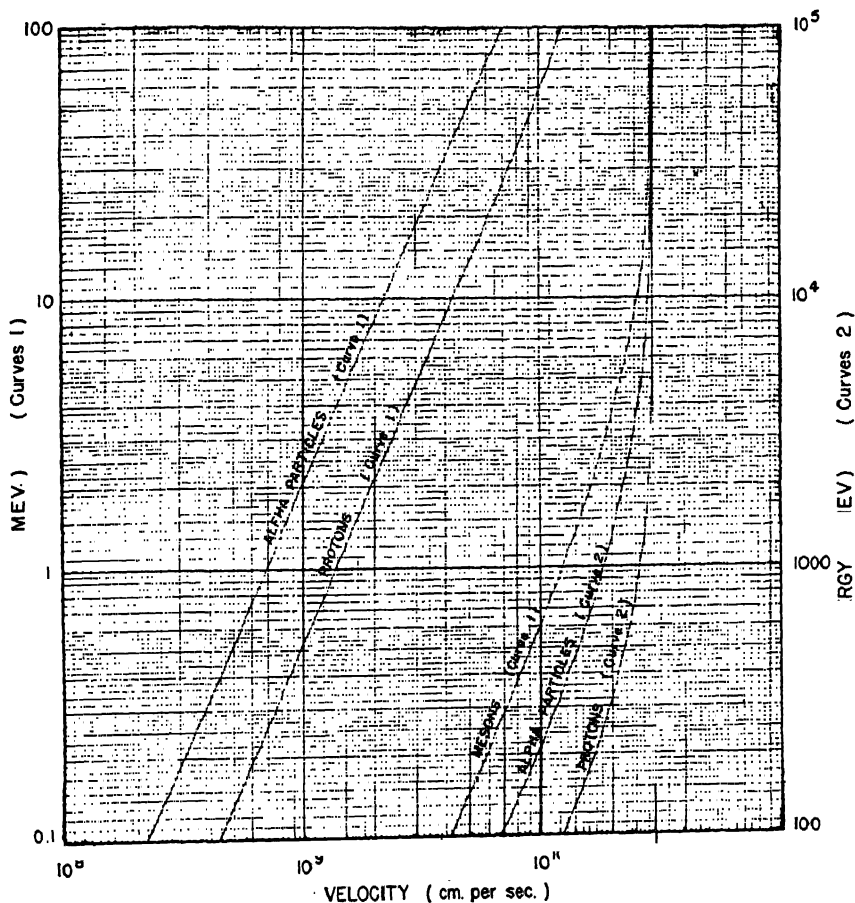
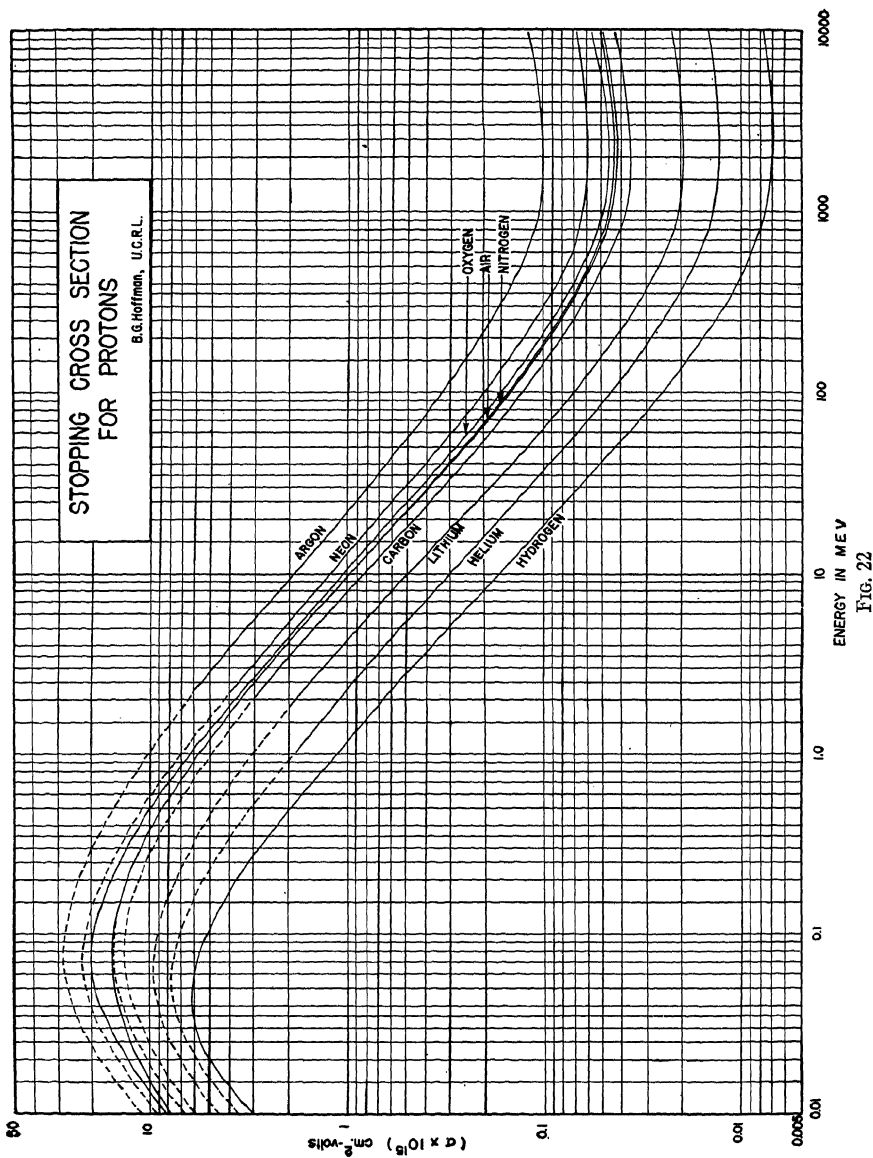


FIG. 21. Energy-velocity curves for alpha particles, protons, and mesons. (Calculated by B. G. Hoffman, University of California, Radiation Laboratory.)



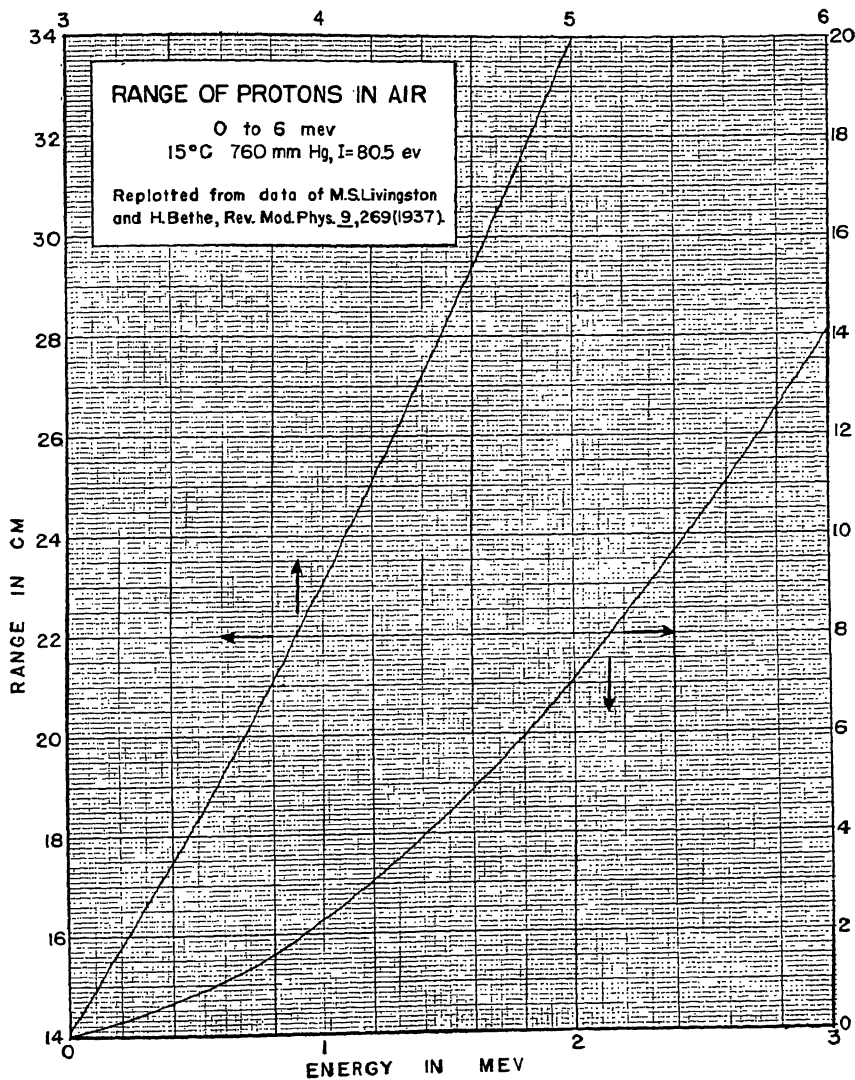


FIG. 23

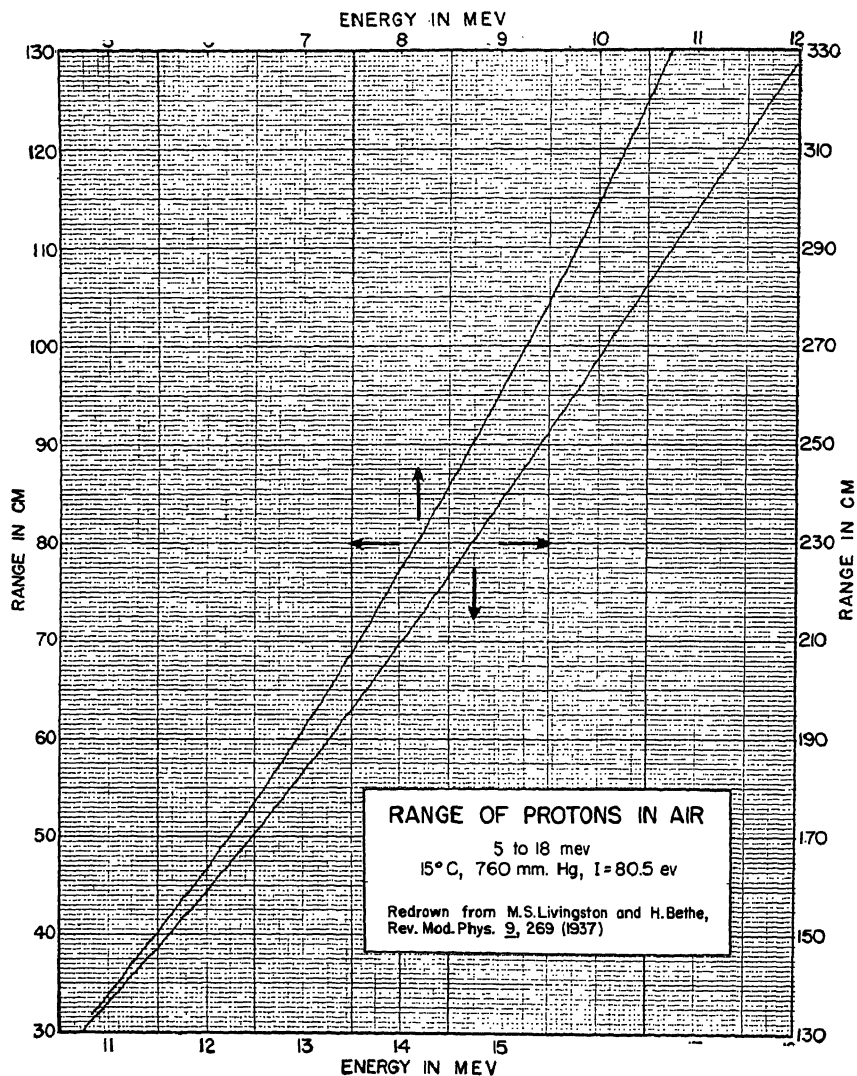


FIG. 24

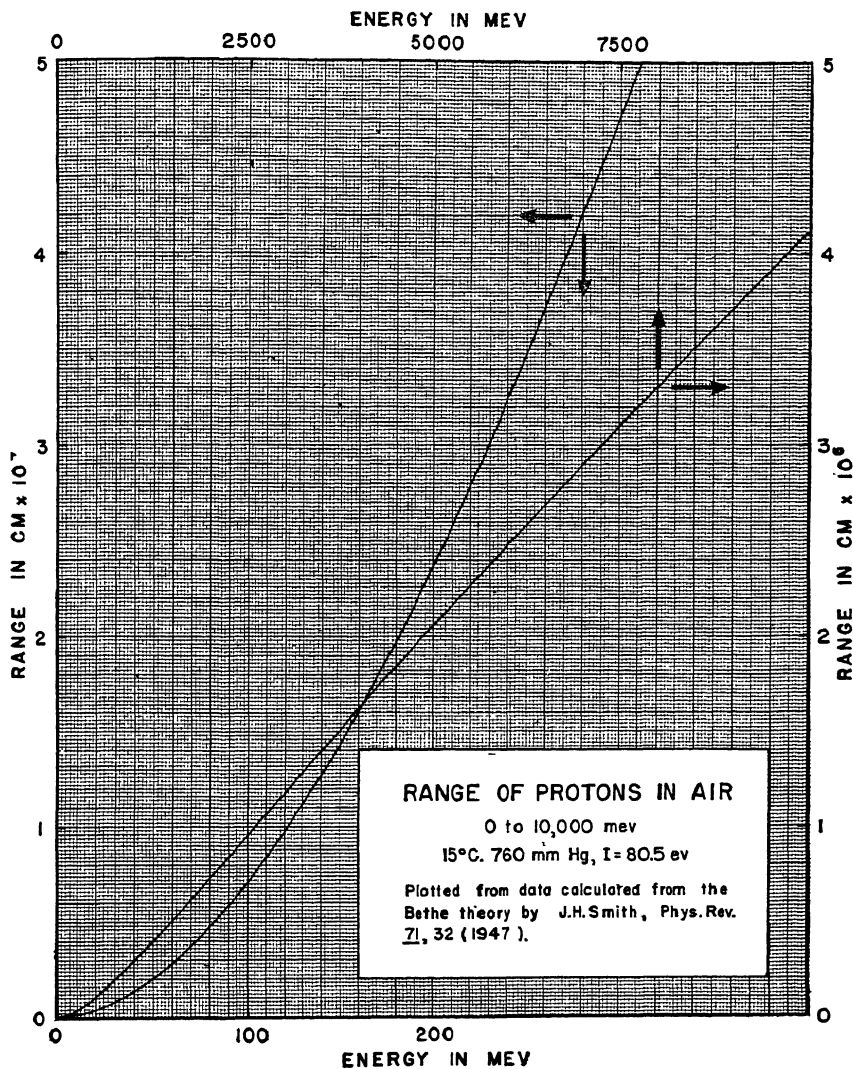


FIG. 25

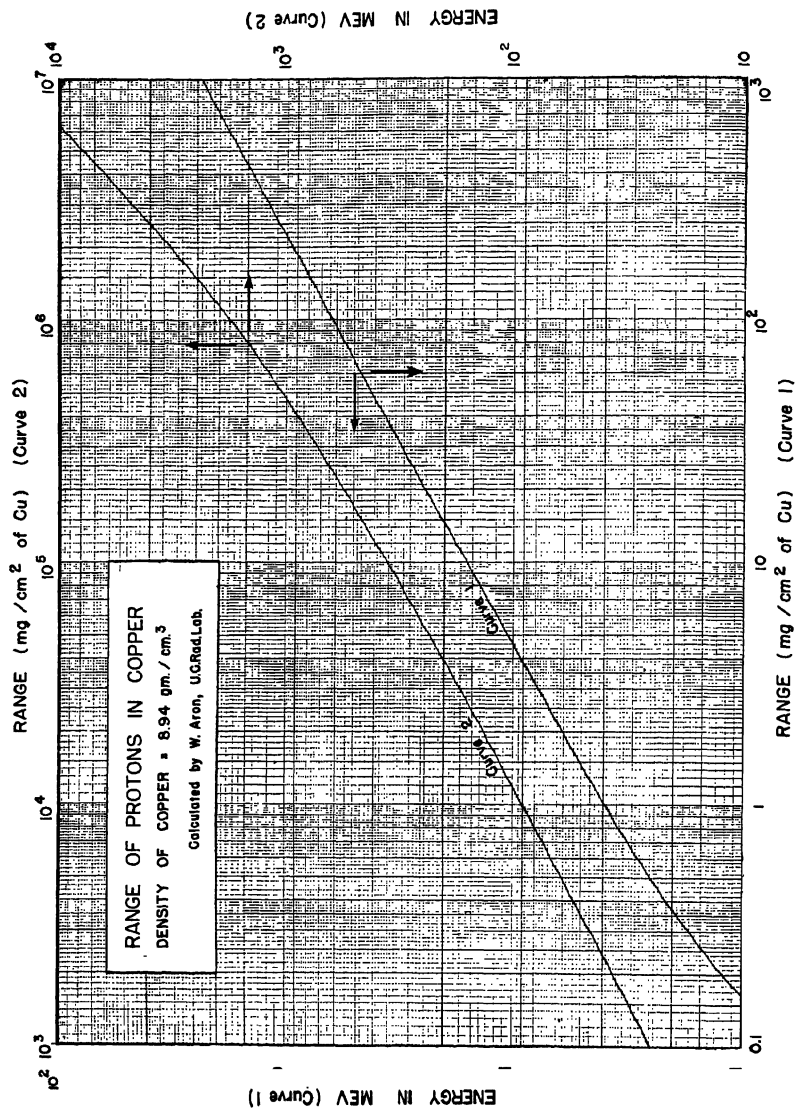


FIG. 26

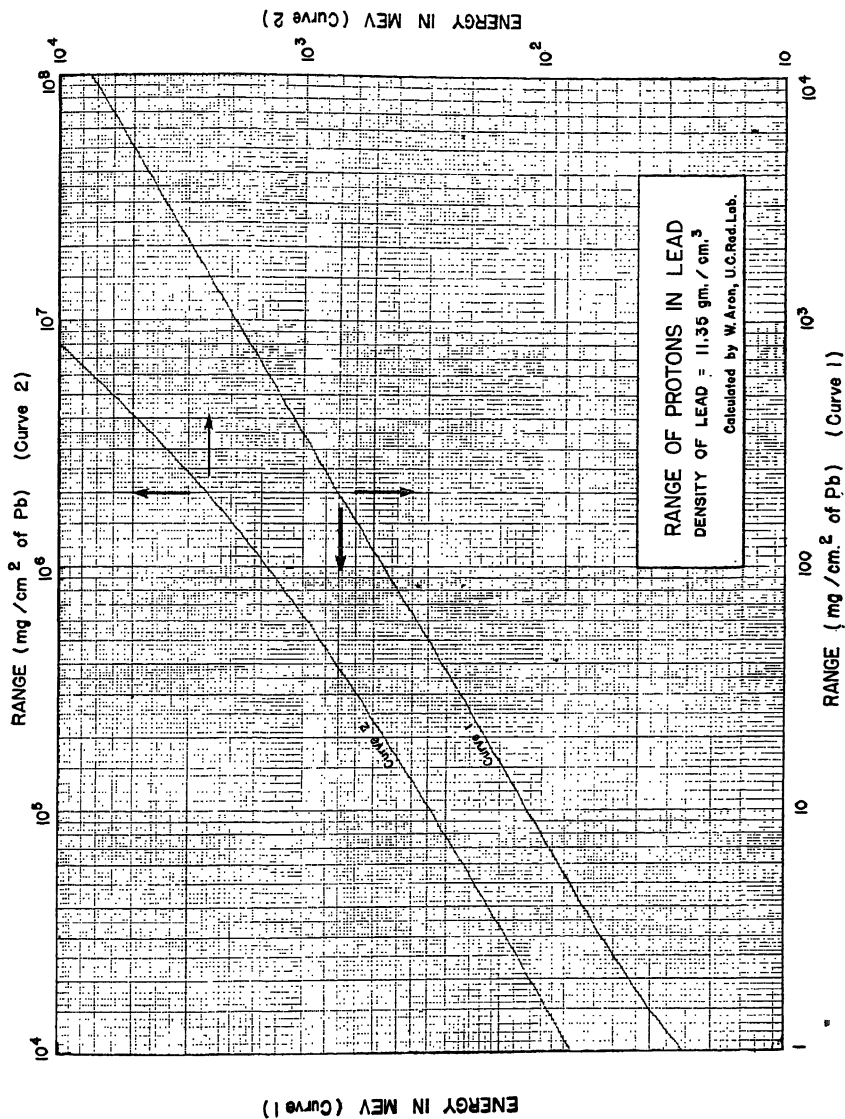


FIG. 27

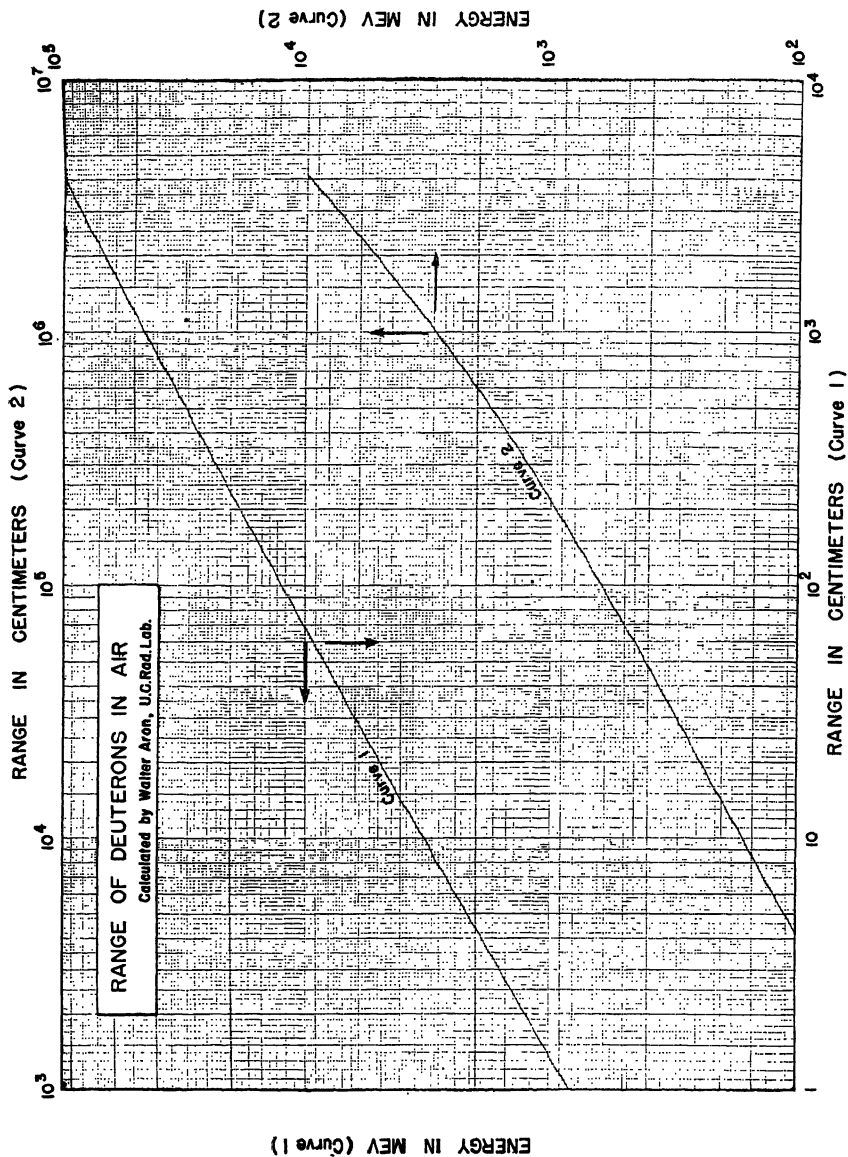


FIG. 28



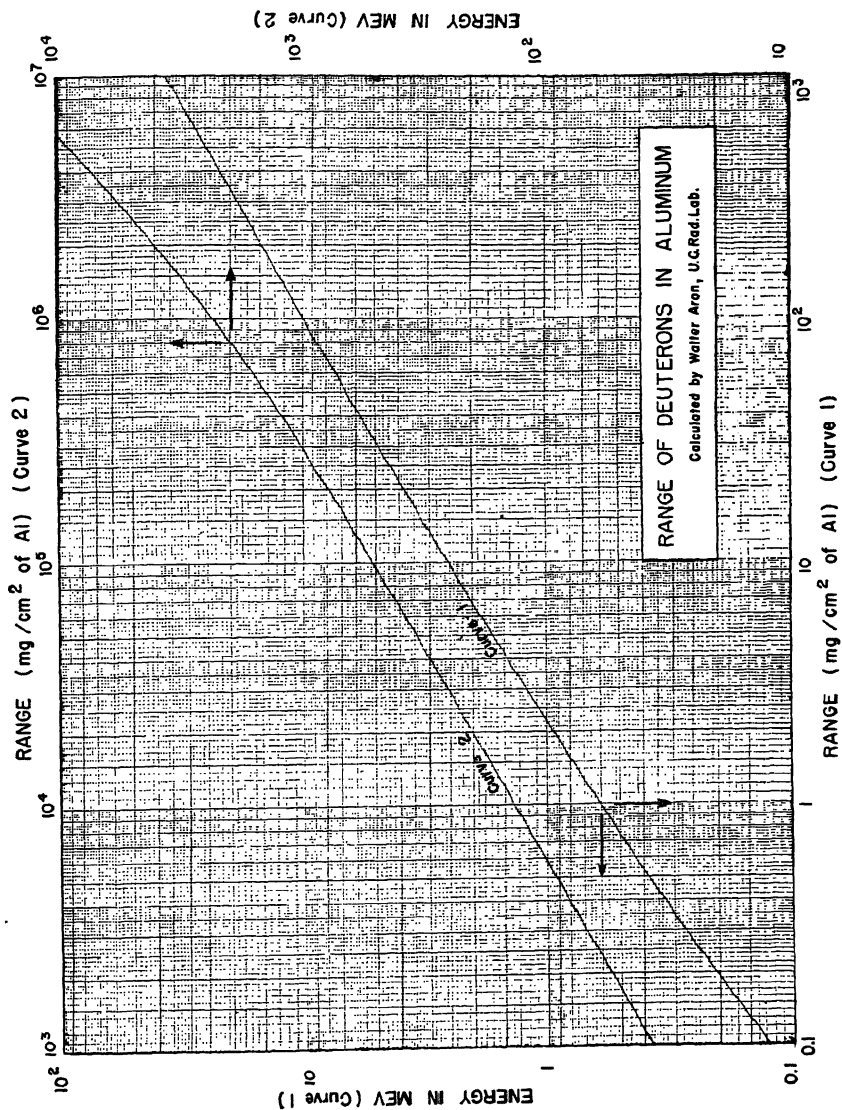


FIG. 29

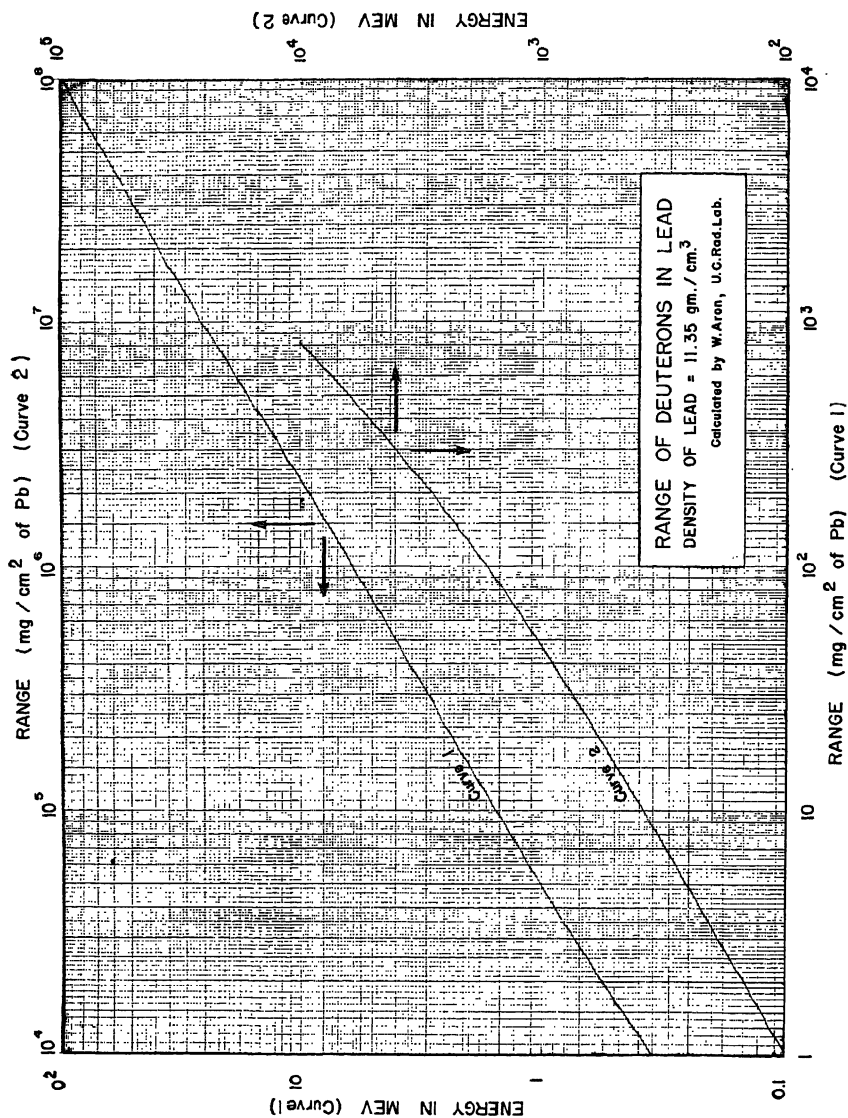


FIG. 30

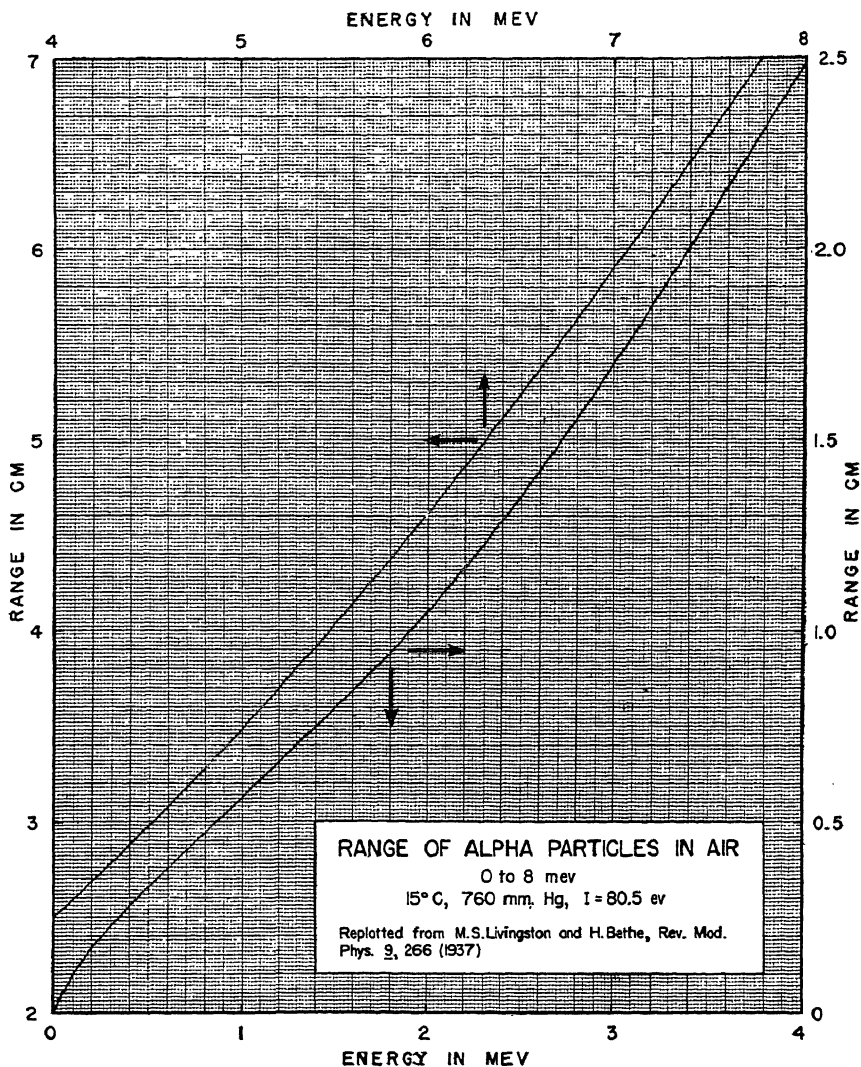


FIG. 31

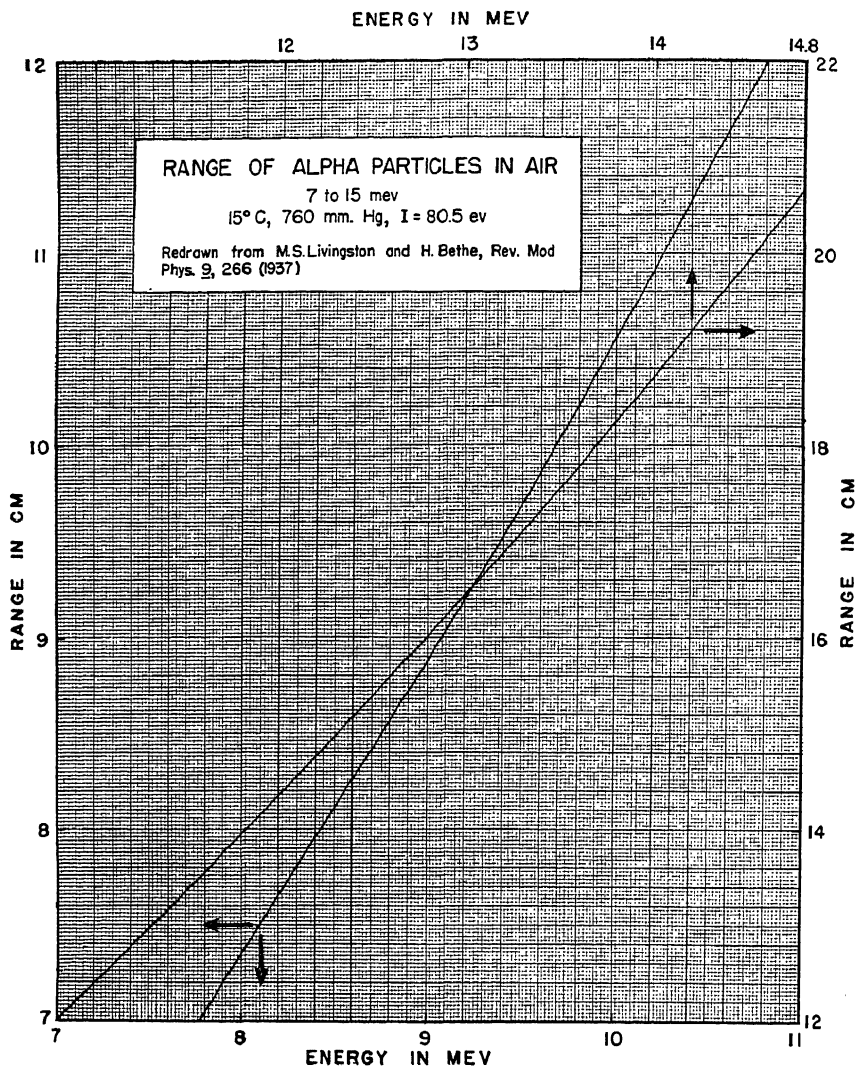


FIG. 32

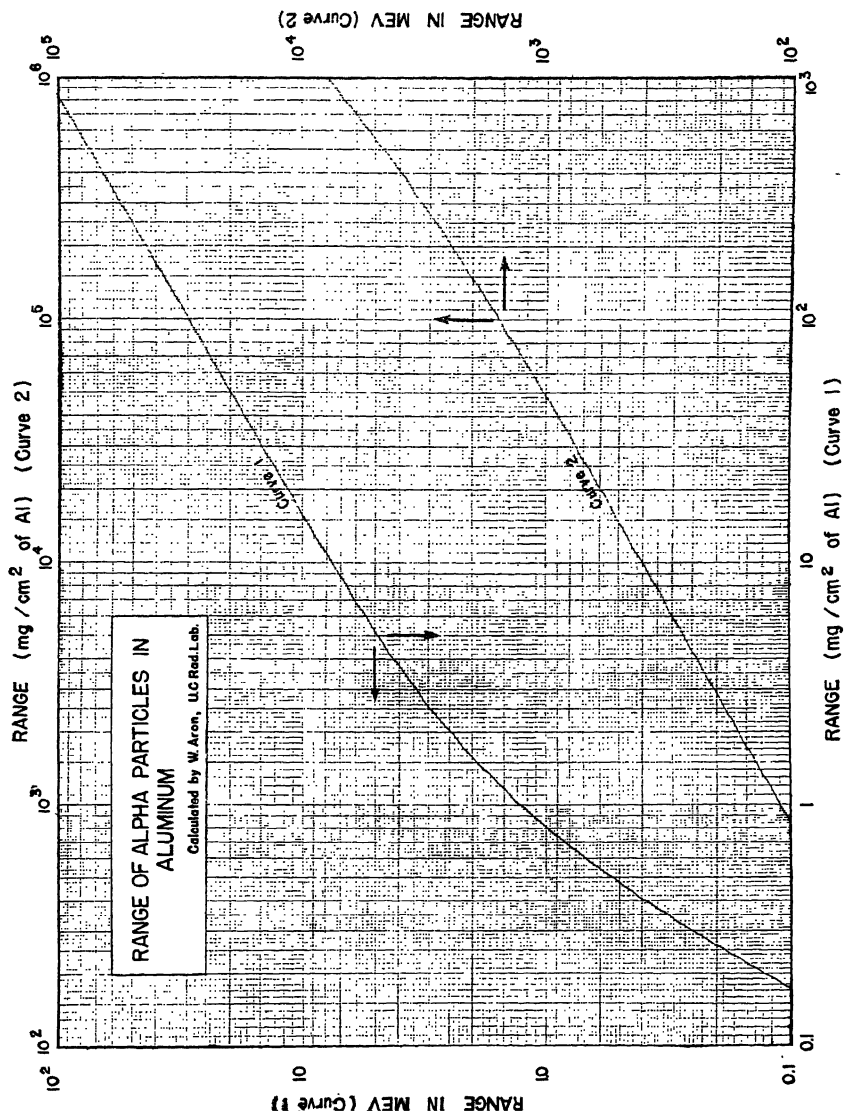
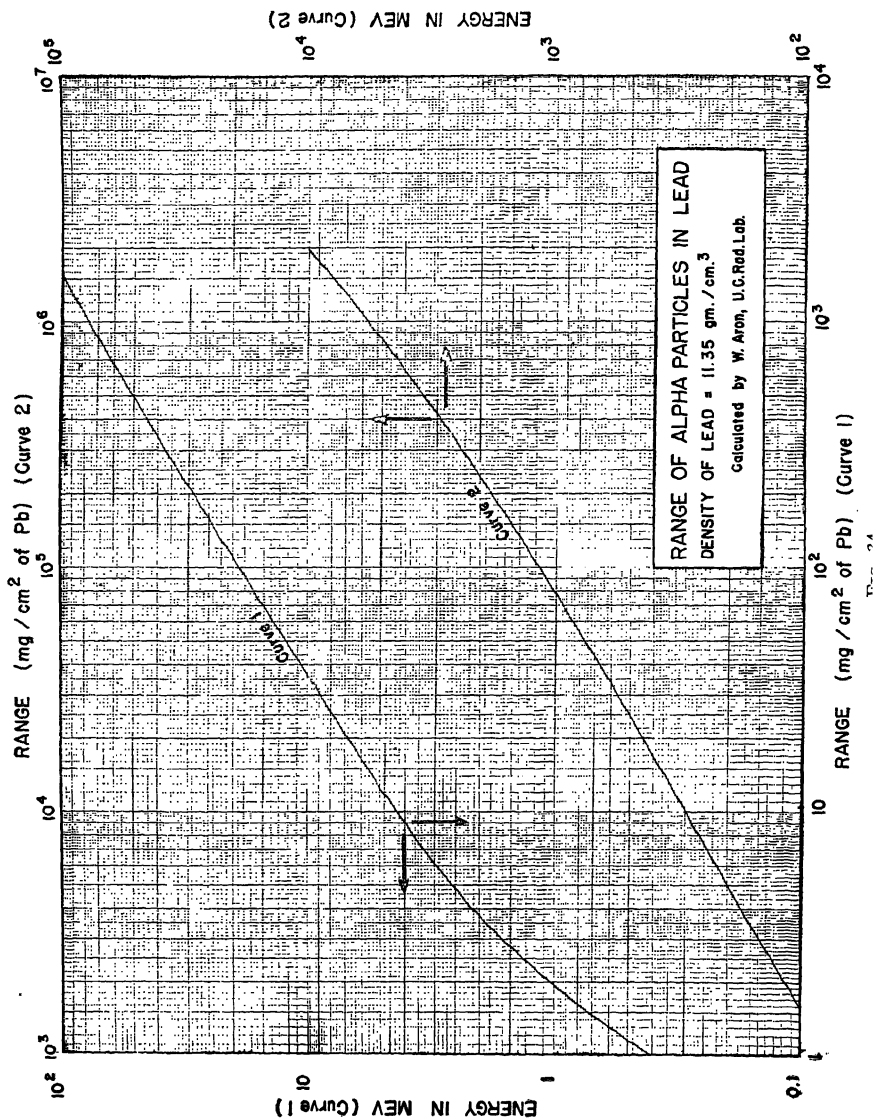


FIG. 33



## CHAPTER 5

### NEUTRONS

#### 5.1. General Properties

Atomic weight [1]:	$= 1.008937 \pm 0.0000075$	
Magnetic moment [2]:	$= -1.91307 \pm 0.0006$	nuclear magnetons
Half-life:	$\sim 20$ min	
Statistics:	Fermi-Dirac	

The absence of charge makes the interaction of neutrons with matter strikingly different from that of charged particles. Interaction with electrons is entirely negligible, and interaction with coulomb fields of nuclei does not occur; hence, neutrons do not lose energy by ionization or radiation. Consequently there does not exist a range-energy relation such as that which describes the behavior of charged particles traversing absorbing media. But by virtue of their electrical neutrality, neutrons of all energies, down to those with nearly zero kinetic energy, have free access to nuclei, with which they readily combine to form unstable compound nuclei. The effects of neutrons on matter, therefore, must be described in terms of the nuclear reactions they produce. More specifically, the essential properties that must be known are the probabilities or cross sections for the various nuclear interactions that may be induced as a function of neutron energy and the kind of nuclei that constitute the matter.

The very marked dependence of neutron processes on energy has led to the universal use of the terms fast, slow, and thermal to indicate broad and rather indefinite ranges of neutron kinetic energy. Fast neutrons are those with energies greater than tens of kev; slow neutrons are those with energies less than this. Included in the latter group are thermal neutrons that possess energies of the order of  $kT$ ,  $\sim 0.02$  ev, where  $k$  is Boltzman's constant and  $T$  is the absolute temperature.

Neutrons from nearly all sources are fast since they are emitted in nuclear reactions with kinetic energies of the order of 1 mev. Consequently slow neutrons can be obtained only by slowing down fast neutrons through elastic collisions, usually with light nuclei. This may be accomplished with extensive volumes of substances such as graphite or water surrounding the fast neutron source. After losing their kinetic energy the neutrons tend to diffuse through matter in much the same way as gases. Under such condi-

tions, when equilibrium is established between the rates of formation and loss of thermal neutrons, the properties of the neutron gas may be described in terms of the spatial density and distribution of kinetic energies, together with associated quantities such as neutron flux, mean free path, and diffusion length.

Associated with each neutron as with all particles is a De Broglie wave for which the wavelength in angstrom units is  $\lambda = h/mv = 0.286E^{-1/2}$  where  $E$  is the kinetic energy in electron volts. Thermal neutrons have wavelengths comparable to interatomic distances, and because of the absence of charge they are diffracted and reflected by crystals in much the same way as x-rays of the same wavelength.

**5.2. Neutron Processes.** In accordance with Heisenberg's uncertainty principle the probability per unit time that an excited nucleus, formed by capture of a neutron, undergoes a transition to a lower excited state is related to the uncertainty in the energy,  $\Delta E$ , of the initial quantum level of the compound nucleus. This is expressed as the probability of a transition per unit time or  $1/\Delta t = \Delta E/\hbar$ , where  $\hbar = \text{Planck's constant}/2\pi$  and  $\Delta t$  is the uncertainty in time which is taken as the mean life of the state. Each quantum state has a most probable energy, its exact resonance energy, but because of the uncertainty principle the energy of the corresponding level in individual nuclei of the same species falls on a distribution curve forming a peak for which the maximum is the most probable value of the level energy. The uncertainty in energy of a particular level, designated by  $\Gamma$  and expressed in electron volts, is taken as the width of the associated resonance peak at one-half its maximum value. It is referred to as the total width of the level. When the width of a level is small, the mean life of the excited state is large.

The lowest excited levels of nuclei are, in general, widely spaced, usually at intervals of the order of several kev. The resonance peaks representing the energies of the levels consequently are well-defined and widely spaced compared with their widths. At higher excitation energies, however, the density of levels increases until, for energies between 5 to 8 mev, the spacing between levels is reduced to only 10 to 20 ev. Discrete resonances then no longer exist; the levels are broad and tend to overlap because of their small separation. Consequently, when nuclei are raised to excited states of high energy, for example by capture of fast neutrons, many levels may be affected which then contribute to the nuclear processes by which the nucleus returns to lower lying levels or to the ground state.

In general the total width  $\Gamma$  is the sum of the partial widths of various processes that may occur in the transition. This includes the emission of gamma rays, neutrons, charged particles and, in the special case of elements beyond actinium, also fission. The total width therefore is  $\Gamma = \Gamma_\gamma + \Gamma_n + \Gamma_\alpha + \Gamma_f$ .



Normally when a slow neutron is captured by a light or medium nucleus, only one such process is highly probable. At high excitation energies, however, several processes may compete with probabilities of roughly the same magnitude.

The kinds of interactions that occur when nuclei are bombarded with neutrons depends, superficially at least, on the kinetic energy of the incident neutrons and on the particular kind of nucleus. In all cases neutron interaction leads to one of the following processes:

1. Elastic scattering. The kinetic energy and momentum of the incident neutron are shared by the recoil neutron and nucleus according to the laws of conservation of energy and momentum; otherwise the struck nucleus is left in its initial state.

2. Radiative capture ( $n, \gamma$ ). The struck nucleus retains the neutron and emits a gamma ray.

3. Neutron emission ( $n, n$ ), ( $n, 2n$ ), etc. After capture of a neutron, one or more neutrons are boiled off.

4. Charged-particle emission ( $n, p$ ), ( $n, d$ ), ( $n, \alpha$ ), etc. After capture of a neutron one or more charged particles, or a combination of particles, are emitted.

5. Fission ( $n, f$ ). A neutron is captured and the compound nucleus splits into two large fragments (see Fission, Chap. 6).

6. Other processes are reported in which very high-energy neutrons ( $\sim 100$  mev) totally disintegrate the nucleus or lead to the emission of a large number of particles (spallation).

All but elastic scattering are capture processes. The incident neutron plus the struck nucleus ( $Z, A$ ) form a compound nucleus ( $Z, A + 1$ ) which instantly ( $\sim 10^{-10}$  to  $10^{-20}$  sec) disintegrates by one of the processes indicated above. The residual nucleus is, except when elastic re-emission of the neutron occurs, radioactive and subsequently decays at a relatively slow rate by emission of those radiations characteristic of radioactive decay.

**5.3. Elastic Scattering of Neutrons.** The precise manner in which neutrons are scattered by nuclei depends in a complicated way on the nuclear forces. Scattering into certain angles may be favored over others (anisotropic scattering), and the cross section usually exhibits strong dependence on neutron energy. At the present time evaluation of such effects of nuclear interaction is possible only from experiment. For many purposes, however, scattering may be considered isotropic in that all angles as measured from the center of mass coordinate system are equally probable.

If scattering is both isotropic and elastic, neutrons are then scattered by nuclei in essentially the same manner as ideal gas molecules. The kinetic energy and momentum of the incident neutron are shared by the recoil neutron and nucleus as required by the conservation laws of energy and

momentum. The struck nucleus remains in other respects unchanged as a result of the collision, and hence no energy is lost to excitation. The kinetic energy transferred to the recoil nucleus is

$$E_M = \frac{4mM \sin^2 \theta/2}{(m + M)^2} E_o = \alpha E_o \quad \text{mev}$$

where  $m$  = mass number of neutron

$M$  = mass number of nucleus

$E_o$  = initial energy of neutron, mev

$\theta$  = deflection angle of neutron from its initial direction with respect to the center of mass

Similarly, the energy lost by the neutron is  $E_n = E_o(1 - \alpha)$ . The greatest amount of energy is lost in a head-on collision where  $\theta = 2\pi$ . If the nucleus is massive, the neutron will reverse its direction but sustain little loss of energy; however, if the struck nucleus is a free proton,  $m \sim M$  and the neutron may in a head-on collision be brought practically to rest in a single collision.

In traversing a scattering medium a fast neutron will make many collisions before it is reduced to thermal velocities. The ratio of the neutron energy after a collision to its initial energy is, on the average, a constant whose magnitude depends on the mass of the scattering nuclei as given by the expression

$$\frac{E_2}{E_1} = e^{-\xi}$$

where

$$\xi = 1 - \frac{(M-1)^2}{2M} \log \frac{M+1}{M-1}$$

$$\approx \frac{2}{M+1} \quad (\text{for heavy nuclei})$$

$E_1$  = initial neutron energy

$E_2$  = final neutron energy

The average number of collisions required to reduce the neutron from energy  $E_o$  to energy  $E$  is then

$$N = \frac{1}{\xi} \log \frac{E_o}{E}$$

If the scatterer is a hydrogenous substance, the average residual kinetic energy of the neutron after a collision with a hydrogen atom is  $1/e$  of its initial value; hence, after  $n$  collisions the residual neutron energy is just  $E = E_o/e^n$ .

The elastic scattering cross section for nuclei must be determined from experiment because of the absence of sufficiently detailed knowledge of nuclear forces. The functional relation of the cross section has, however,

been established [4]. For isotropic scattering, it may be expressed as the sum of two distinct processes. The first arises from potential scattering for which the cross section is identical to that obtained for the collision of two hard spheres and is just equal to the total effective surface area of the nucleus. The second or resonance term becomes important when the energy of the incident neutron is nearly equal to that of a quantum state of the nucleus. Resonance scattering may then make a large contribution to the cross section. Breit and Wigner [3] have shown that when the De Broglie wavelength of the neutron is large compared to the nuclear radius and the levels are widely spaced so that only one level is affected, the cross section near resonance can be represented by an expression similar to that for the dispersion of light. The total cross section, therefore, may be written as the sum of potential scattering and the Breit-Wigner "one-level" formula [3,4].

$$\sigma_s = 4\pi R^2 + \frac{\pi}{2} \left( 1 \pm \frac{1}{2i+1} \right) \lambda_r \Gamma_n \frac{4R(E - E_r) + \lambda_r \Gamma_n}{(E - E_r)^2 + \frac{\Gamma^2}{4}} \quad \text{cm}^2$$

where  $R$  = effective nuclear radius

$E$  = neutron energy

$E_r$  = resonance energy

$\lambda_r$  = neutron wavelength at resonance  $(=h^2/8\pi^2\mu E)^{1/2}$

$\mu$  = reduced mass  $= mM/(m+M)$

$\Gamma_n$  = neutron width of level

$\Gamma$  = total level width

$i$  = angular momentum of nucleus before collision, + sign if spin of resonance level is  $i + \frac{1}{2}$ ; - sign if  $i - \frac{1}{2}$ . If  $i = 0$  (even-even nuclei) + sign is used

The shape of the curve for the second term near resonance is similar to the dispersion curve for light. A maximum occurs near exact resonance where  $\sigma_{\max} = 4\pi R^2 + 2\pi\lambda_r^2\Gamma_n/\Gamma$ , and a minimum value is found at

$$E_r - E_{\min} = \lambda_r \Gamma_n / 2R$$

where  $\sigma_{\min} = 2\pi R^2$ .

The relative magnitude of resonance and potential scattering is made apparent by the ratio of their cross sections at exact resonance

$$\frac{\sigma_r}{\sigma_p} \approx \frac{1}{2} \left( \frac{\lambda_r \Gamma_n}{R\Gamma} \right)^2$$

For most nuclei the neutron width  $\Gamma_n$  is a small fraction ( $\sim 10^{-3}$ ) of the total width  $\Gamma$ , and since  $\lambda_r \approx R$ , the quantity  $\lambda_r \Gamma_n / R\Gamma \ll 1$ . Hence, in most cases resonance scattering is unimportant compared with potential scattering. There are however a few elements such as silver for which

$\Gamma_n \approx \Gamma$ , and the  $\sigma$  at exact resonance is then very much greater than off-resonance values. In general, since potential scattering is the more important factor, the elastic scattering cross section appears to be relatively independent of energy.

Elastic scattering is not easily observed in most nuclei since it is usually obscured by more competitive capture processes. Only in the light elements, excepting  $\text{Li}^6$  and  $\text{B}^{10}$ , and a few elements of medium weight are scattering cross sections greater than for capture. Hydrogen is the most important scattering nucleus since it causes the greatest average reduction in energy per collision and exhibits the largest elastic scattering cross section. In the neutron energy range from 1 ev to about 10 kev, the scattering cross section of hydrogen is nearly constant, with a value of about  $\sigma_s = 21$  barns. At lower energies it increases rapidly ( $\sigma_s \approx 85$  barns for thermal neutrons, see Figs. 35 and 36).

**5.4. Interaction of Slow Neutrons with Nuclei.** The interaction of neutrons with nuclei was first treated by Breit and Wigner for the case in which only one nuclear resonance level was important in the process [3]. This was later extended to the more general problem by Bethe and Placzek [5] and others [6,7,8,9].

If only one level is important in the interaction process because of the wide spacing of levels in the energy range to which a slow or thermal neutron can excite the nucleus, then the cross section for the formation of a compound nucleus and the emission of a particle (or gamma ray) of the kind  $q$  is given by the one-level formula in the form

$$\sigma_a = \frac{\pi}{2} \left( 1 \pm \frac{1}{2i + 1} \right) \frac{\lambda \lambda_r \Gamma_n \Gamma_q}{(E - E_r)^2 + \Gamma^2/4} \quad \text{cm}^2$$

where  $\lambda$  = De Broglie wavelength of neutron

$\lambda_r$  = De Broglie wavelength at resonance

$E$  = neutron energy

$E_r$  = resonance energy

$\Gamma_n$  = neutron width of level

$\Gamma_q$  = level width for emitted particle (or gamma ray)

$\Gamma$  = total width for all processes

The total width  $\Gamma$  includes all possible processes summed over their respective widths. This includes neutron emission, gamma-ray and charged-particle emission as well as fission. Since the neutron width varies as  $E^{1/2}$ , the total width can be written in the form

$$\Gamma = \Gamma_n \left( \frac{E}{E_r} \right)^{1/2} + \sum \Gamma_q \quad \text{ev}$$

Ordinarily, for slow neutrons only one kind of particle has a high probability for emission and the contribution of other processes to  $\Gamma$  is negligible. Furthermore, the emission of a neutron (inelastic scattering) is important only when simple capture with gamma emission is unlikely and when the emission of a charged particle is energetically impossible because of the potential barrier. But usually,  $\Gamma_n \ll \Gamma_q$  as seen from the fact that the observed ratio for these two widths is  $\sim 10^{-3}$ . The total width can, therefore, in many instances be set equal to the particle width:  $\Gamma = \Gamma_q$ . In individual cases, the relative magnitude of the neutron width can be determined experimentally from the cross section at exact resonance since the cross section is then directly proportional to the ratio of the neutron to particle width.

$$(\sigma_n)_r = \frac{\pi}{2} \left( 1 \pm \frac{1}{2i + 1} \right) \lambda_r^2 \frac{\Gamma_n}{\Gamma} \quad \text{cm}^2$$

From the one-level formula for elastic scattering and the similar formula above for capture, the ratio of the two cross sections gives the relative probability at resonance for elastic scattering (neglecting potential scattering) with respect to a competing capture process (with particle or gamma-ray emission).

$$\frac{\sigma_s}{\sigma_q} = \frac{\Gamma_n}{\Gamma_q} \left( \frac{E}{E_r} \right)^{3/2}$$

This ratio is greater than unity for most of the light elements (except  $\text{Li}^6$  and  $\text{B}^{10}$ ) and in general is less than unity for elements of medium atomic weight (except iron, nickel, and copper).

The emission width  $\Gamma_q$  for charged particles is the product of the particle width without the potential barrier,  $\Gamma'_q$ , and the penetrability  $P$  of the barrier, or  $\Gamma = \Gamma'_q P_q$ , where  $P_q = e^{-f}$  and  $f$  is a function of the height and width of the barrier. Because of the factor  $P$ , charged-particle emission in all but the lightest elements and in the special case of fission is extremely improbable under slow neutron bombardment. Proton emission is known only for  $\text{N}^{14}$  although it is energetically possible for  $\text{B}^{10}$  [4]. Alpha-particle emission is observed only in  $\text{Li}^6$  and  $\text{B}^{10}$  which have very large cross sections, about 3,000 and 900 barns, respectively. Without a more exact and detailed model for the nucleus, the most probable process and its width must be determined experimentally. In general, it will include any one of the processes  $(n, n)$ ,  $(n, p)$ ,  $(n, \alpha)$ ,  $(n, \gamma)$  and fission.

If the energy of the neutron is small compared to the first resonance level, or if it is small compared to the width of the first level, *i.e.*, if  $E \ll E_r$  or  $E \ll \Gamma$ , then the resonance terms become unimportant and the cross section for capture varies directly with  $\lambda_r$ , or as  $1/v$ , where  $v$  is the neutron velocity. This is observed most clearly in rhodium, indium and iridium for thermal neutrons, and in  $\text{B}^{10}$  for energies as high as  $\sim 0.1$  mev.

**5.5. Interaction of Fast Neutrons with Nuclei.** The interaction of fast neutrons with nuclei involves a single nuclear energy level only when the level spacing is comparable to the total neutron energy. This is sometimes the case for light elements and for energies up to about 1 mev, corresponding to excitation energies of the order of 5 to 8 mev. In medium and heavy nuclei and in light nuclei for high neutron energies, more than one level is excited in the formation of the compound nucleus and in the subsequent emission of radiation (gamma, neutron, or charged particle). The one-level formula is then not valid. When the levels are closely spaced, an average cross section for a particular process must be obtained by summing the one-level formula over all states of the final nucleus. If it can be assumed that the average level spacing is small, *i.e.*, the level density is high, instead of taking a sum of the contributions the averaging can be performed by integrating over an energy range large as compared to the level spacing but small compared to the neutron kinetic energy. The average cross section for the capture of fast neutrons and emission of a particular radiation  $q$  is then given by

$$\sigma = \pi R^2 \xi \frac{\Gamma_q}{\Gamma} \quad \text{cm}^2$$

where  $R$  = nuclear radius

$\Gamma_q$  = particle, radiation, or fission width

$\Gamma$  = total width for all processes

$\xi$  = sticking probability

Further, the total average cross section for any and all processes,  $q$ , which can occur,  $(n, n)$ ,  $(n, \gamma)$ ,  $(n, \text{charged particle})$ , fission, etc., is found by summing over the width for all processes.

$$\sigma = \pi R^2 \xi \quad \text{cm}^2$$

The sticking probability  $\xi$  as used here is defined as the probability that a neutron which strikes a nucleus will stick, *i.e.*, be absorbed to form a compound nucleus. For high excitation energies where the nuclear level spacing is small, this is roughly  $\xi = \Gamma_n/D$ , where  $\Gamma_n$  is the neutron width and  $D$  is the average level spacing. For high-energy neutrons, and particularly in medium and heavy elements, the sticking probability is nearly unity. For medium energies ( $\sim 1$  mev) it appears to be of the order of 0.1 and for low energies, considerably less [4]. At very high energies, therefore, the total cross section is just equal to the geometric cross-sectional area of the nucleus,  $\pi R^2$  ( $\sim 10^{-25}$  cm<sup>2</sup>). This, necessarily, corresponds to energies for which the neutron wavelength is less than the nuclear radius.

Brief descriptions of the separate processes are given below.

*a. Inelastic Scattering*  $(n, n)$ . The emitted neutron is ejected with less kinetic energy than the incident neutron. Only in the event that the

nucleus is left in the ground state after the collision are the energies of the two neutrons equal; in which case the process is indistinguishable from elastic scattering. For inelastic scattering, the difference between the energies of the initial and emitted neutrons is contributed to the excitation of the residual nucleus and is subsequently emitted as gamma radiation. The direction taken by the ejected neutron is purely random since, in effect, the nucleus does not remember the original direction of incident neutron. Inelastically scattered neutrons, therefore, are observed to be emitted uniformly over a sphere. Also it is to be noted that the energy and momentum laws for the collision of hard spherical particles do not hold since part of the energy is lost to excitation.

If the incident neutron energy is less than the lowest nuclear level, only elastic scattering can occur. On the other hand, when the energy is sufficiently high to excite many levels, a neutron may be emitted by any one of many possible level transitions and the observed energy distribution of the neutrons is approximately Maxwellian [8].

The observed  $(n, n)$  cross section for fast neutrons is found, in many substances, to be a large fraction of the total cross section for all possible processes.

*b. Radiative Capture  $(n, \gamma)$ .* Since inelastic scattering is known to occur in most instances with an appreciable probability, if it is assumed that this and radiative capture are the only possible process in a particular nucleus, the cross section for  $(n, \gamma)$  is

$$\sigma_{(n,\gamma)} = \pi R^2 \xi \frac{\Gamma_\gamma}{\Gamma_n + \Gamma_\gamma} \quad \text{cm}^2$$

The gamma width  $\Gamma_\gamma$  does not appear to be a sensitive function of the neutron energy and has a value ranging from 0.01 to 1.0 ev for slow neutrons which is assumed to be roughly the same for neutrons of medium energy (10 to 500 kev). Bethe [4] gives for the average capture cross section the expression

$$\sigma_\gamma = 2 \times 10^{-22} E^{-1/2} \quad \text{cm}^2$$

At very high energies, the radiative capture cross section becomes exceedingly small and multiple-neutron and charged-particle emission becomes far more probable.

*c. Multiple-neutron Emission  $(n, 2n)$ ,  $(n, 3n)$ , . . .* More than one neutron is emitted provided that the nucleus is left in a sufficiently excited state following emission of the first neutron; i.e., if  $E_b < E_i - E_e$ , where  $E_b$  is the binding energy of second neutron in the nucleus,  $E_i$  is the energy of incident neutron, and  $E_e$  is the energy of first emitted neutron.

Assuming that the energy distribution of the emitted neutrons can be

approximated by a Maxwell distribution for a temperature  $T$  because of the large number of levels affected at high energies, the cross section for emission of two neutrons is given by the expression

$$\sigma_{(n,2n)} = \pi R^2 \xi \left[ 1 - \left( 1 + \frac{E_n - E_b}{T} \right) e^{\frac{E_b - E_n}{T}} \right] \quad \text{cm}^2$$

where  $T$  = temperature correlated to observed energy distribution,

$$T = 2(5E_n/A)^{1/2} \text{ mev}$$

$\xi$  = sticking probability for neutrons of high energy ( $> 1.0$  mev)

At high energies  $E_n - E_b \gg T$ , and the  $(n, 2n)$  process becomes more probable than  $(n, n)$ . The process involving the emission of more than two neutrons becomes increasingly more important at very high energies, *i.e.*, when  $E_i \gg E_b$ .

*d. Charged-particle Emission*  $(n, p)$ ,  $(n, d)$ ,  $(n, \alpha)$ , . . . . Reactions leading to the emission of charged particles are probable only when the incident neutron energy is sufficiently high so that the available energy is great enough to allow a charged particle to pass over the electrostatic potential barrier. Assuming the average cross section for high energies to be of the form

$$\sigma_q = \pi R^2 \xi \frac{\Gamma_q}{T} \quad \text{cm}^2$$

the charged particle width is the product of the emission width without the barrier  $\Gamma'_q$  and the penetrability factor  $P = e^{-J}$ . Because of the large numbers of levels excited by neutrons with energies high enough to result in charged-particle emission, the energy distribution of the emitted particles is continuous and probably roughly Maxwellian.

Ordinarily, the cross section for charged-particle emission is small because of the strong competition from the more favorable  $(n, n)$  process or at very high energies, from  $(n, 2n)$ ,  $(n, 3n)$ ; etc.

*e. Fission* (see Fission, Chap. 6).

*f. Other Processes.* At energies of the order of 100 mev and greater, neutron bombardment frequently leads to the emission of great numbers of particles and probably complete disintegration of certain nuclei. Such processes are still under investigation at the present time (184-in. synchrocyclotron at the Radiation Laboratory of the University of California).

**5.6. Neutron Diffusion.** For practical purposes neutrons are obtained either by the reactions  $(\alpha, n)$  and  $(\gamma, n)$  from mixtures of a radioactive isotope such as radium, and a "target" material such as beryllium, or from appropriate targets bombarded with heavy particles in high-energy accelerators. However, all such sources provide only fast neutrons, and when slow neutrons are required it is necessary to slow down the fast neutrons with appropriate



forms of moderators. If the source (or the fast neutron beam from a cyclotron) is enclosed with materials such as water or paraffin for which the ratio of the scattering to capture cross section is large, the fast neutrons are rapidly reduced to thermal energies by elastic collisions.

Once neutrons have been reduced to thermal velocities they continue for the remainder of their lives to diffuse through the medium. On the average they gain in subsequent collisions as much energy as they lose, and their average velocity depends only on the temperature of the medium. Ultimately each neutron is lost by capture, by decay into a proton and an electron, or by diffusion out of the medium. When equilibrium is reached, the rate of formation of thermal neutrons just equals the rate of loss by all three processes, and the neutron density throughout the medium remains constant with time although not in space. If the diffusing medium is a hydrogenous substance such as paraffin, most of the scattering nuclei have the same mass number as the neutron and diffusion resembles in some respects the self-diffusion of gases. Substances containing heavy nuclei, on the other hand, allow very small momentum transfer per collision, and the neutrons scatter elastically as from a solid, immovable object. The conditions here are more like the diffusion of an electron gas in a conductor.

In the stationary state, characterized by equal rates of thermal neutron loss and gain, the energy distribution of neutrons is Maxwellian except for a high-energy tail which varies as  $E^{-3/2}$ . For the Maxwellian region the number of neutrons with energies lying in the interval  $E$  to  $E + dE$  is

$$N dE = \frac{2Q\tau E^{1/2}}{(\pi k^3 T^3)^{1/2}} e^{-E/kT} dE$$

where  $k$  = Boltzman constant

$T$  = absolute temperature

$Q$  = number of neutrons produced per sec

$\tau$  = mean life time for capture

The factor  $Q\tau$  is just the total number of neutrons with all energies. The average velocity is  $\bar{v} = (8kT/m\pi)^{1/2}$ , where  $m$  is the mass of the neutron. At room temperature,  $20^\circ\text{C}$ ,  $\bar{v}$  is about 2.5 km per sec.

Assuming that all neutrons within a diffusing medium possess thermal velocities, or more exactly, have a Maxwellian distribution for a given temperature, the neutron density in neutrons per cubic centimeter may then be described by a diffusion equation in much the same way as for gases. It is essentially an expression of the law of conservation of neutrons stating that the rate of change in the number of neutrons per cubic centimeter in an element of volume  $dV$  at the point  $(x, y, z)$  and time  $t$  equals the number of neutrons produced per second plus the divergence of the neutron flux through  $dV$  minus the number absorbed per cubic centimeter per second.

$$\frac{dn}{dt} = \text{div} (D \text{ grad } n) + q - \frac{n}{\tau}$$

where  $D$  = diffusion constant

$q$  = source strength; number of thermal neutrons produced per unit time as a function of position

$\tau$  = mean life of neutron before capture

In the steady state  $dn/dt = 0$ ; the density distribution does not change with time. Furthermore, if the medium is homogeneous and isotropic, the diffusion equation then reduces to

$$D\nabla^2 n + q - \frac{n}{\tau} = 0$$

or

$$\nabla^2 n + \frac{q}{D} - \frac{n}{\tau D} = 0$$

The diffusion constant  $D$  is found from ordinary kinetic theory to be given by  $D = \lambda_{tr}v/3$ , where  $\lambda_{tr}$  is the transport mean free path and  $v$  is the neutron velocity. This relation is found to hold for neutron diffusion provided that the scattering is isotropic, scattering and absorption cross sections are constant, and the rate of change of  $n$  is small within a distance of one  $\lambda_{tr}$ . The quantity  $\lambda_{tr}$  is the mean distance a neutron travels through the medium in the direction of its initial motion after a great number of collisions. This is related to the transport cross section by  $\lambda_{tr} = 1/N\sigma_{tr}$ , where  $N$  is the number of nuclei per cubic centimeter. The quantity  $\sigma_{tr}$  is, in turn, defined by  $\sigma_{tr} = \sigma_s(1 - \overline{\cos \theta})$ , where  $\sigma_s$  is the scattering cross section and  $\overline{\cos \theta}$  is the average value of the cosine of the angle of deflection from the initial direction in each collision. If the scattering is strictly isotropic, i.e., all angles are equally probable, then  $\overline{\cos \theta} = 0$ ,  $\sigma_{tr} = \sigma_s$  and  $\lambda_{tr} = \lambda$ , where  $\lambda$  is the mean free path or  $\lambda = 1/N\sigma$ . The value of  $\overline{\cos \theta}$  for anisotropic scattering depends on both the mechanism of scattering and the mass number of the scattering nuclei. The first effect must be determined empirically, but the latter effect leads to  $\sigma_{tr} = \sigma_s(1 - 2/3M)$ , where  $M$  is the mass number of the scattering nucleus. In the case of hydrogen, for example, scattering is predominantly forward since a neutron cannot be scattered more than 90 deg in a single collision. The effect in hydrogen is large, whereas in heavy elements it is entirely negligible.

In some media the absorption cross section  $\sigma_a$  is comparable to the scattering cross section and cannot be neglected. In such instances a more accurate expression for the diffusion constant, taking  $\sigma_a$  into account, is

$$D = 3N\sigma_{tr}[1 - (\frac{2}{3})(\sigma_a/\sigma_{tr})]$$

The source strength  $q$ , the number of thermal neutrons formed per cubic centimeter per unit time, is in general a function of the coordinates  $x, y, z$ . Most sources provide only fast neutrons which require 20 to 30 collisions to be reduced to thermal velocities. Hence, for a point source of fast neutrons the virtual source of slow neutrons is some function of the radial distance. In many instances, however, slow neutrons are obtained either from a moderator, used to slow down neutrons from a cyclotron, or as a collimated beam from a nuclear reactor. The source can then often be regarded as a plane with a strength of  $n_0 \bar{v}$ , where  $n_0$  is the number of neutrons per cubic centimeter of beam and  $\bar{v}$  is the average velocity.

The mean life  $\tau$  of a neutron with respect to capture by a nucleus is related to the absorption cross section by the expression  $\tau = 1/N\sigma_a \bar{v}$ .

The quantity  $\tau D$  in the diffusion equation is referred to as the diffusion length  $L$ . It often makes its appearance in solutions of the equation in terms of the form  $e^{-x/L}$ , where  $x$  is the distance from the source. If scattering is isotropic and if  $\sigma_a \ll \sigma_s$ , the diffusion length is

$$L = \frac{1}{N(3\sigma_s\sigma_a)^{1/2}} \quad \text{cm}$$

If scattering is not isotropic and  $\sigma_a$  is not small compared with  $\sigma_s$ , a more exact expression for  $L$  is

$$L = \frac{1}{N[1 - (2\sigma_a/5\sigma_s)](3\sigma_s\sigma_s)^{1/2}} \quad \text{cm}$$

Examples of solutions of the diffusion equation may be illustrated by two different and simple diffusion geometries which are of practical interest.

The first is that of a point source of neutrons placed at the center of a sphere of scattering material of radius  $R$ . The density of neutrons at any radial distance  $r$  from the center is

$$n = \frac{2Q}{2\pi\lambda\bar{v}r} \frac{e^{R/L}}{(e^{2R/L} + 1)} \sinh \frac{R-r}{L}$$

where  $Q$  is the source strength in neutrons produced per unit time and  $\bar{v}$  is the average velocity.

The second geometry considered here is a semi-infinite medium with a plane boundary. It is assumed that the source is also a plane with a strength of  $Q$  neutrons per square centimeter per second and placed deep within the medium. It is apparent that this geometry may be used to obtain a rough approximation of the distribution in a thick slab of material into which a beam of slow neutrons is directed. From the plane side opposite that which the beam enters the neutron density is a linearly increasing function of

depth. At a depth  $x$  measured along the normal to the boundary the neutron density is

$$n = Q \left( \frac{x}{D} + \frac{2}{v} \right)$$

At the surface,  $n$  has the value  $2Q/v$  inside the medium and the value zero just outside. If the straight line representing  $n$  within the medium is extended beyond the boundary, it goes to zero at a distance  $d = 0.71\lambda$  from the surface, a distance referred to as the extrapolated end point.

The steady-state distribution of neutrons depends, as indicated by the diffusion equation, on the geometry of the medium and the disposition of the sources. The density distribution is, however, strongly affected also by the presence of strongly absorbing materials placed within the medium and by the character of material bounding the diffusing medium. Absorbing materials such as indium or cadmium foils depress the neutron density in the region near the absorber since they behave as sinks, absorbing all thermal neutrons that enter. Most such substances, however, are nearly transparent to fast neutrons. Those elements which possess large capture cross sections, some of which are commonly used for neutron detectors and shields, are given in Table 19.

TABLE 19. THERMAL NEUTRON ABSORBERS  
(Approximate total cross section for neutrons of 0.024 ev energy)

Element	Cross section, barns	Element	Cross section, barns
Li	70	Sm	6,000
B	750	Eu	4,700
Rh	150	Gd	46,000
Ag	65	Dy	1,200
Cd	2,500	Re	100
In	200	Ir	430
Au	110	Hg	400

Neutrons that reach the bounding surface of the diffusing medium are ordinarily lost. If, however, a second nonabsorbing medium is placed around the first, some of the neutrons that cross the boundary are subsequently scattered back, thus raising the neutron density. The effect of a reflector, which may be any substance including the original diffusing medium, is expressed in terms of its albedo  $\gamma$ , defined as the ratio of the number of neutrons that flow back in across the boundary to the number that flow out. For a spherical diffusing medium of radius  $R$ , surrounded by a reflector with a thickness very much greater than the mean free path of the neutrons,  $\gamma$  has the value

$$\gamma = \frac{1 - \frac{2}{3}[(\lambda/R) + (\lambda/L)]}{1 + \frac{2}{3}[(\lambda/R) + (\lambda/L)]}$$

where  $\lambda$  = mean free path in reflector =  $1/N\sigma$

$L$  = diffusion length

The value of  $\gamma$  increases with  $R$  and for  $R \rightarrow \infty$  becomes the albedo for a plane boundary

$$\gamma = \frac{1 - (2\lambda/3L)}{1 + (2\lambda/3L)}$$

Material such as concrete is particularly useful for reflectors and also serves as a barrier around moderators to reduce the health hazard to personnel working in the vicinity.

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TABLE 20. THERMAL NEUTRON ACTIVATION CROSS SECTIONS

The following table is a list of thermal neutron activation cross sections measured and reported by L. Seren, H. N. Friedlander, and S. H. Turkel, *Phys. Rev.*, **72**, 888 (1947). Cross sections were determined by measuring the absolute beta emission (or positron and K capture) from thin foils irradiated with thermal neutrons. Assuming that one beta particle is emitted per neutron captured, the thermal neutron cross section  $\sigma$  was calculated from  $u = n\sigma N$ , where  $u$  = number of neutrons captured per unit time,  $n$  = thermal neutron flux, and  $N$  = number of atoms of detector. The probable error of most values is 20 per cent; those with a probable error of 10 per cent are indicated by \* and those with a probable error of 40 per cent by †.

Isotope			Half-life	Isotopic cross section, barns	Natural atom cross section, barns
Z	El.	A			
8	O	18	31 s	$2.2 \times 10^{-4}$	$4. \times 10^{-7}$
9	F	19	12 s	0.0094	0.0094
11	Na	23	14.8 h	0.63	0.63
12	Mg	26	10.2 m	0.048	0.0054
13	Al	27	2.4 m	0.21	0.21
14	Si	30	170 m	0.116	0.00485
15	P	31	14.3 d	0.23	0.23
16	S	34	87.1 d	0.26	0.011
17	Cl	35	$2 \times 10^6$ y	0.169	0.13
		37	37 m	0.56	0.137
19	K	41	12.4 h	1.0	0.067
20	Ca	40	8.5 d	$< 0.000125^\dagger$	$< 0.00012^\dagger$
		44	180 d	0.63	0.013
		48	30 m	0.55	0.00105
		48	150 m	0.025	0.00039
21	Sc	45	85 d	22.	22.
22	Ti	50	6 m	0.141	0.0075
		50	72 d	0.039	0.0021
23	V	51	3.9 m	4.50	4.50
24	Cr	50	26.5 d	11.†	0.50†
		54	1.3 h	$\sim 0.0061$	$\sim 0.00014$
25	Mn	55	2.59 h	10.7	10.7
26	Fe	58	47 d	0.36	0.0010
27	Co	59	10.7 m	0.66	0.66
		59	5.3 y	21.7	21.7
28	Ni	64	2.6 h	1.96	0.0173
29	Cu	63	12.8 h	2.82	2.0
		65	5 m	1.82	0.56
30	Zn	64	250 d	0.51	0.26
		68	57 m	1.09	0.19
		68	13.8 h	0.31	0.054
31	Ga	69	20 m	1.40	0.855
		71	14.1 h	3.36	1.30

TABLE 20. THERMAL NEUTRON ACTIVATION CROSS SECTIONS—(Continued)

Isotope			Half-life	Isotopic cross section, barns	Natural atom cross section, barns
Z	El.	A			
32	Ge	70	40 h	0.073	0.0155
		70	11 d	$\sim 0.45\dagger$	$\sim 0.095\dagger$
		74	89 m	0.38	0.14
		76	12 h	0.085	0.0055
33	As	75	26.8 h	4.2	4.2
34	Se	74	115 d	22.†	0.2†
		78, 80	19 m	.....	0.23
		78, 80	57 m	.....	0.017
		82	30 m	0.060	0.0056
35	Br	79	18 m	8.1	4.1
		79	4.4 h	2.76	1.39
		81	34 h	2.25	1.11
37	Rb	85	19.5 d	0.72	0.52
		87	17.5 m	0.122	0.033
38	Sr	86	2.7 h	1.29	0.127
		88	55 d	0.0050	0.00415
39	Y	89	60 h	1.24	1.24
40	Zr	92	63 d	0.33	0.073
		94	17.0 h	0.053	0.009
		96	6 m	$\sim 1.07\dagger$	$\sim 0.016\dagger$
41	Cb	93	6.6 m	$\sim 1.0\dagger$	$\sim 1.0\dagger$
42	Mo	100	19 m	0.475	0.044
		98	67 h	0.415	0.10
		92?	7 h	.....	0.001†
44	Ru	102, 104	4 h	.....	0.122
		?	37 h	.....	0.15
		?	40 d	.....	0.37
45	Rh	103	44 s	137.*	137.*
		103	4.2 m	11.6*	11.6*
46	Pd	108	13 h	11.2	3.0
		110	26 m	0.39	0.0525
47	Ag	107	2.3 m	44.3	23.
		109	22 s	97.	46.6
		109	225 d	2.3	1.1
48	Cd	114	2.5 d	1.1	0.30
		114	43 d	0.14	0.040
		116	3.75 h	1.4	0.10
		?	2 m	.....	0.05
49	In	113	48 d	56.*	2.52*
		115	13 s	51.8*	49.5*
		115	54 m	144.6*	138.*
50	Sn	112	105 d	$\sim 1.1^*$	$\sim 0.012\dagger$
		124	9 m	0.574	0.039
		< 125	40 m	.....	0.0142

TABLE 20. THERMAL NEUTRON ACTIVATION CROSS SECTIONS—(Continued)

Isotope			Half-life	Isotopic cross section, barns	Natural atom cross section, barns
Z	El.	A			
50	Sn	< 125	26 h	.....	0.072
		< 125	10 d	.....	0.009
		< 125	400 d	.....	0.018
51	Sb	121	2.8 d	6.8	3.8
		123	60 d	2.5	1.1
52	Te	126	9.3 h	0.78	0.15
		126	90 d	0.073	0.014
		128	72 m	0.133	0.0436
		128	32 d	0.0154	0.00504
		130	25 m	0.222	0.0735
		130	30 h	< 0.008†	< 0.003†
53	I	127	25 m	6.25	6.25
55	Cs	133	3 h	0.016	1.016
		133	1.7 y	25.6	25.6
56	Ba	138	86 m	0.511	0.367
57	La	139	40 h	8.4	8.4
59	Pr	141	19.3 h	10.1	10.1
62	Sm	?	21 m	.....	1.10
		152	46 h	138.	35.8
		?	60 d	.....	< 0.008†
63	Eu	151	9.2 h	1,380.	681.
		151, 152	5-8 y	.....	390.
64	Gd	?	9.5 h	.....	2.3
		?	20 h	.....	~ 0.9†
		?	8.6 d	.....	~ 0.6†
		?	160 d	.....	< 0.25†
65	Tb	159	3.9 h	10.7	10.7
66	Dy	164	140 m	2,620.	725.
		164	1.25 m	120.	33.
67	Ho	165	30 h	59.6	59.6
69	Tm	169	105 d	106.	106.
71	Lu	175, 176	3.4 h	.....	15.9
		176	6.6 d	3,640.	91.0
72	Hf	180	46 d	10.0	3.5
73	Ta	?	16.2 m	.....	0.034
		181	117 d	20.6	20.6
74	W	184	77 d	2.12	0.64
		186	24.1 h	34.2	10.2
75	Re	185	90 h	101.	38.5
		187	18 h	75.3	46.5
76	Os	190	30 h	2.50	0.66
		192	17 d	5.34	2.19
77	Ir	191	1.5 m	260.	100.
		191	70 d	1,000.	388.



TABLE 20. THERMAL NEUTRON ACTIVATION CROSS SECTIONS—(Continued)

Isotope			Half-life	Isotopic cross section, barns	Natural atom cross section, barns
El.					
77	Ir	193	20.7 h	128.	79.0
78	Pt	196	18 h	1.1	0.30
78	Pt	196	3.3 d	4.5	1.20
		198	31 m	3.92	0.292
79	Au	197	2.7 d	96.4	96.4
80	Hg	204	5.5 m	0.34	0.0428
		202, 204	51.5 d	.....	0.725
81	Tl	203	4.23 m	0.273	0.079
		205	3.5 y	3.1	2.2
83	Bi	209	5.0 d	0.015	0.015

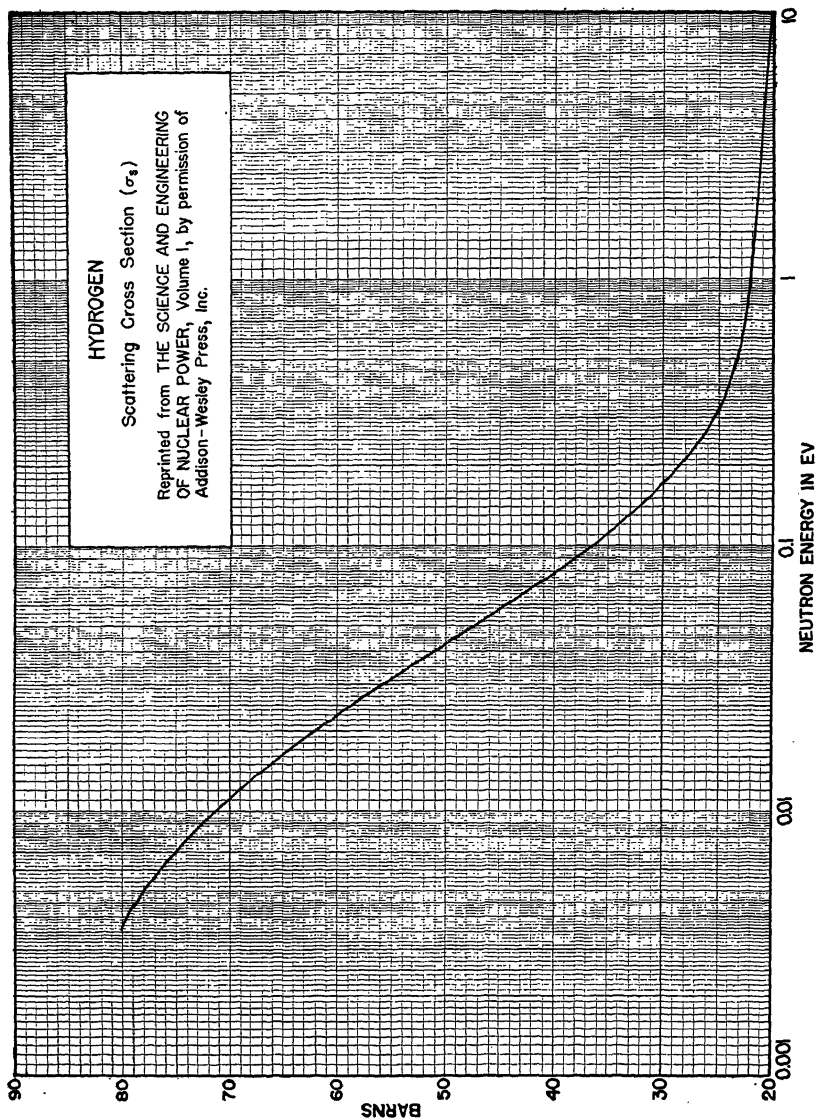


FIG. 35

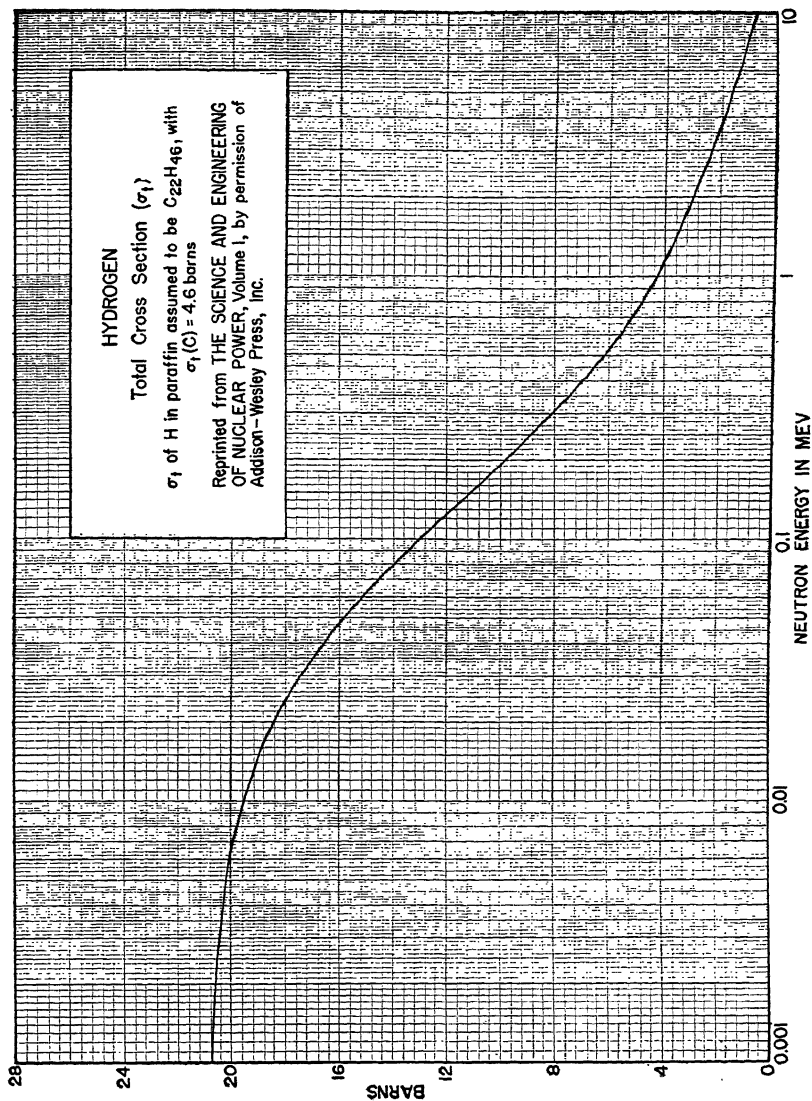


FIG. 36

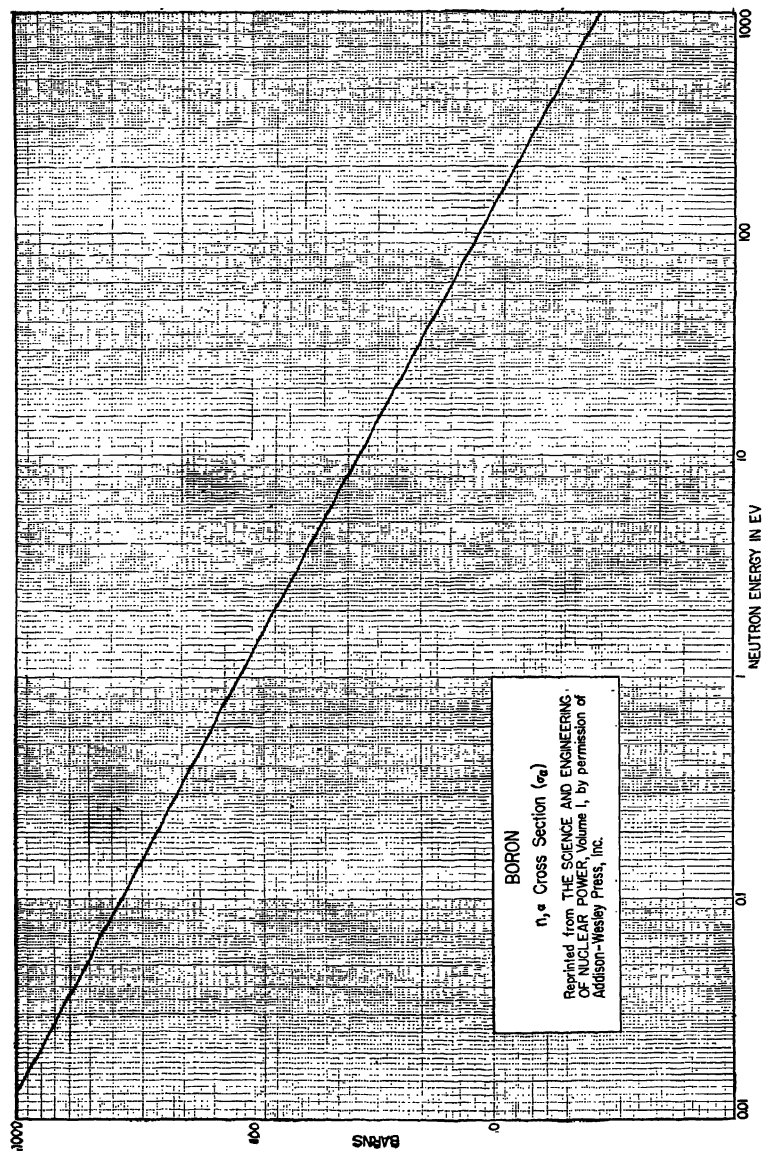


FIG. 37

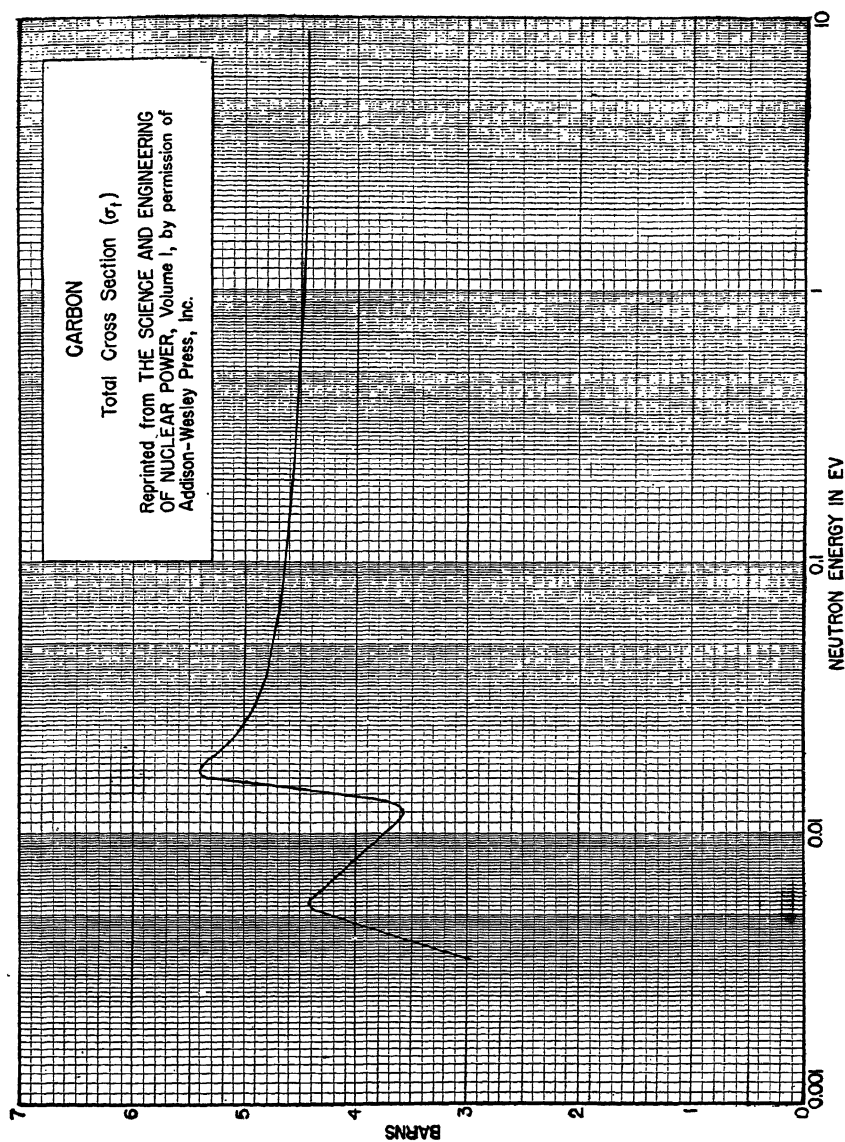


FIG. 38

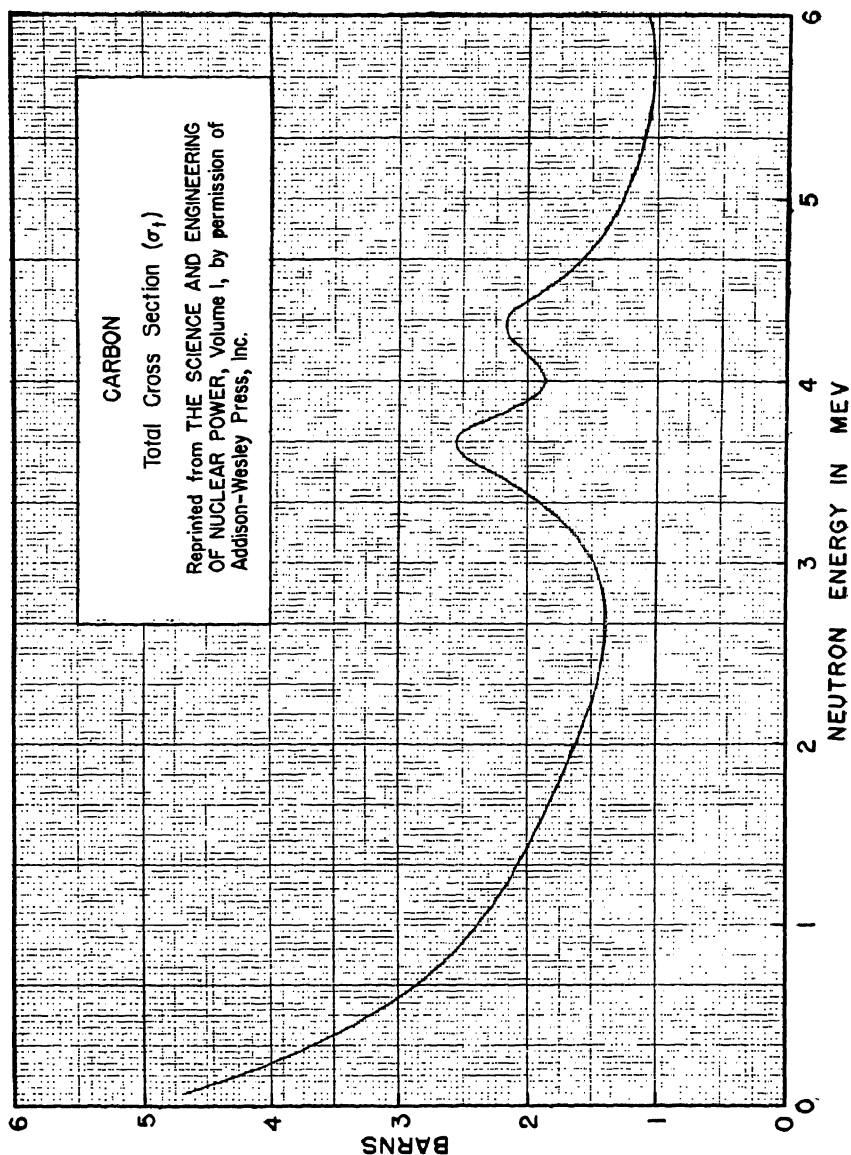


FIG. 39

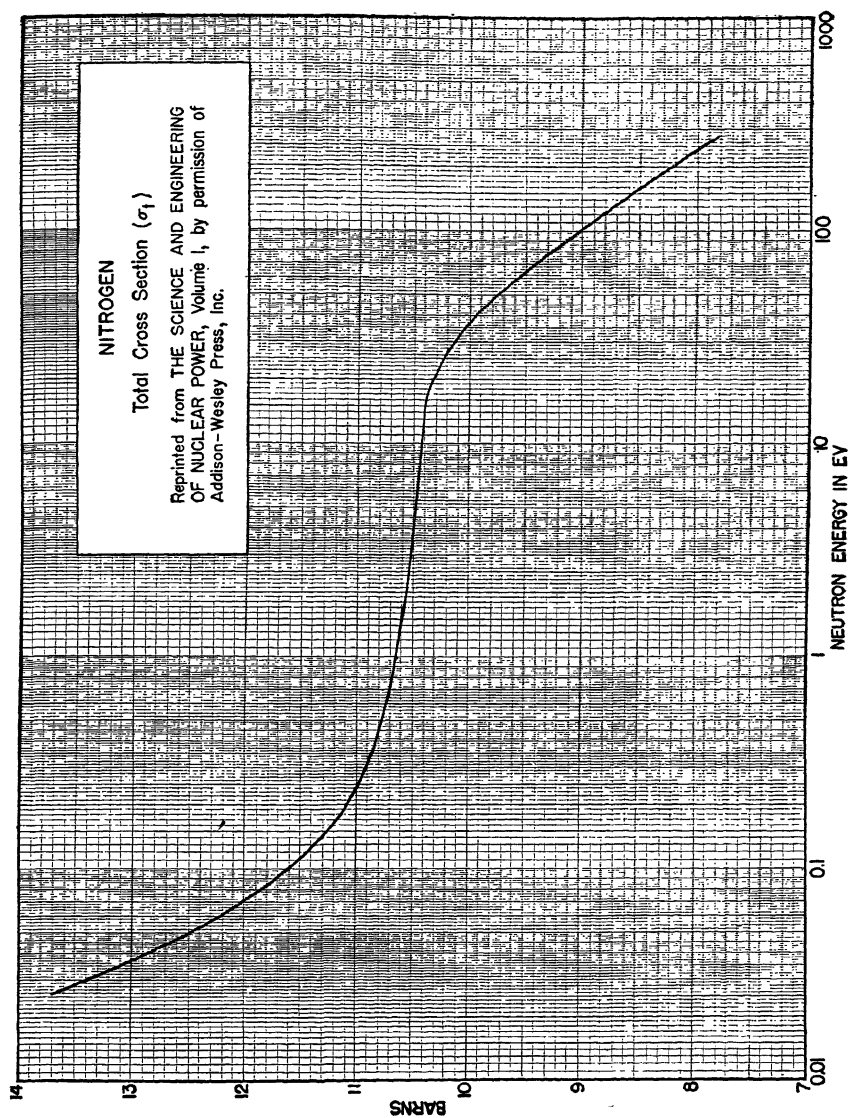


FIG. 40

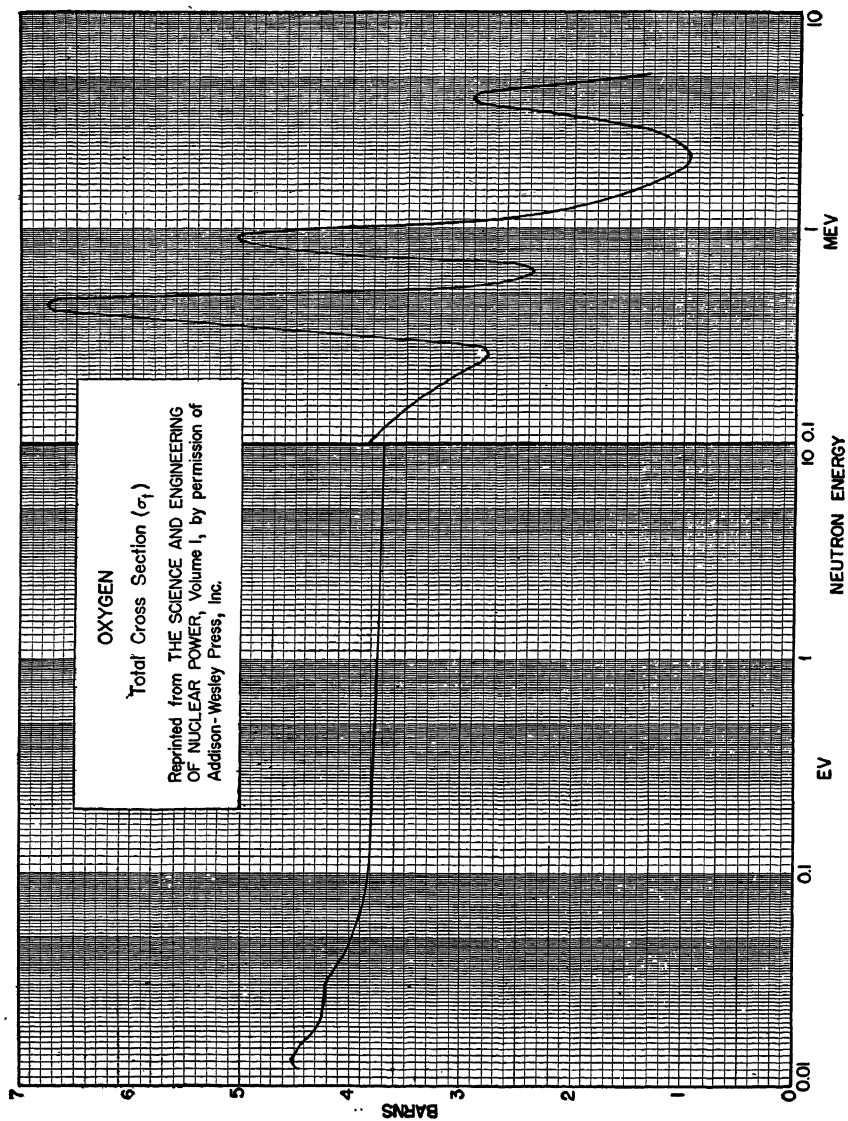


FIG. 41



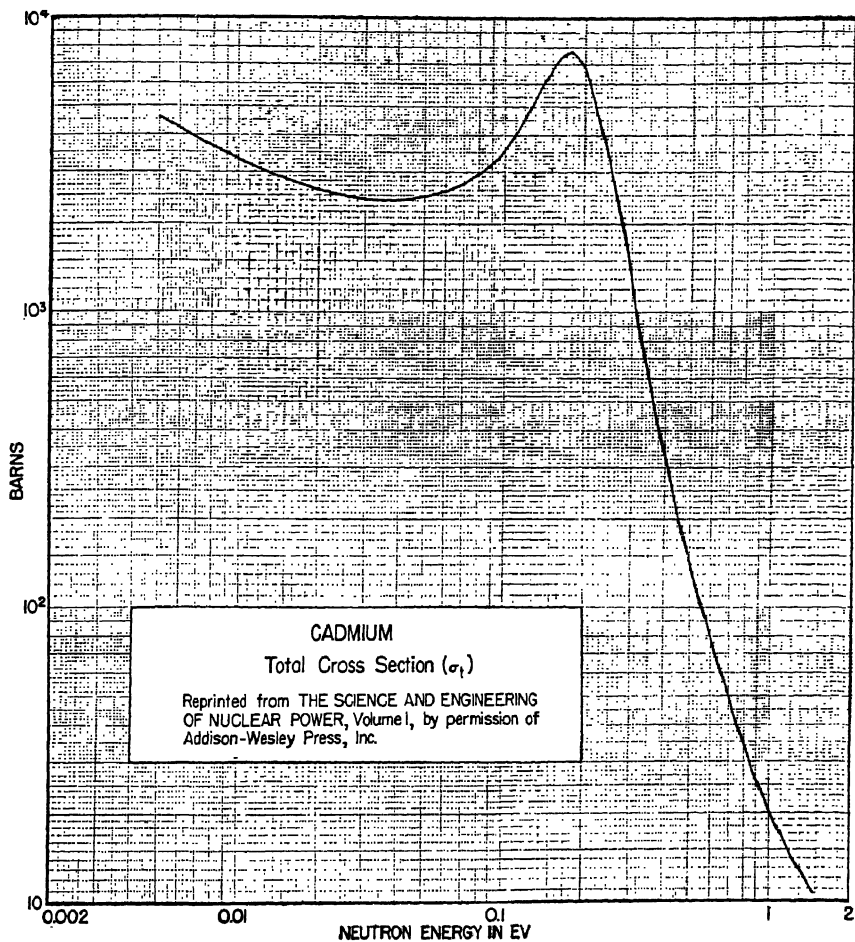


FIG. 42

## CHAPTER 6

### FISSION

**6.1. Mechanism of Fission.** The discovery by Hahn and Strassmann [1] that elements of medium atomic weight were formed from uranium bombarded with neutrons came as the culmination of an unexplained phenomenon encountered in the investigations of Fermi, Meitner, Hahn, Strassmann, Curie, and Savitch. Meitner and Frisch [2] named the process "fission" from its similarity to the rupture of a sphere of charged incompressible fluid as a result of deformations induced by some external influence. The analogy of the "liquid drop" had already been applied in principle to heavy nuclei by Bohr and Kalckar [3], and the general theory of liquid-sphere dynamics had been developed in detail for its application to stellar dynamics. Following the discovery of fission, Bohr and Wheeler [4] extended the liquid-drop analogy into a theory of fission with marked success in its detailed agreement with observations of fission processes.

The aggregate of elementary particles in heavy nuclei, according to this model, forms the equivalent of a spherical drop of incompressible liquid with a volume proportional to the number of particles or atomic weight and an effective surface tension arising from the short-range attractive fields between the particles. The surface tension is compensated for in part by the repulsive electrostatic field of the uniform distribution of proton charge  $eZ$  throughout the nucleus. Under the influence of an external force, the nucleus may be excited into one of many possible modes of vibration, producing deformations in the spherical form similar to the corresponding vibrations of a liquid sphere. The stability of the drop to these small deformations depends on the relative strengths of the surface tension and the counteracting electrostatic field. As the total number of particles in stable nuclei increases, a limiting size is attained for which the compensation of the cohesive nuclear force by the electrostatic field is complete and the drop is then unstable to spontaneous fission. Nuclei only slightly smaller than the limiting size, although exceedingly stable against spontaneous fission, will divide when sufficiently excited. A small deformation accompanying excitation allows a redistribution of charge to the surface regions of smaller curvature, thus giving the repulsive field an advantage over the surface tension in regions of smaller curvature. A sufficiently large deformation therefore leads to complete division, and the separated fragments recoil, because of their similar charges,

with a total kinetic energy of about 160 mev. The high excitation energy of the fragments following fission is subsequently lost by the emission of prompt (instantaneous) neutrons and gamma rays and through radioactive chains leading finally to known stable nuclei.

**6.2. Fission Probability.** The probability of fission induced by an incident neutron depends (1) on the probability of neutron capture to form a compound nucleus and (2) on the relative probabilities for deexcitation of the compound nucleus by radiation, neutron emission, and fission. Both the neutron binding energy and the critical energy for fission in the heaviest nuclei lie in the energy range of 5 to 7 mev. When the binding energy plus the kinetic energy of the captured neutron is less than the critical fission

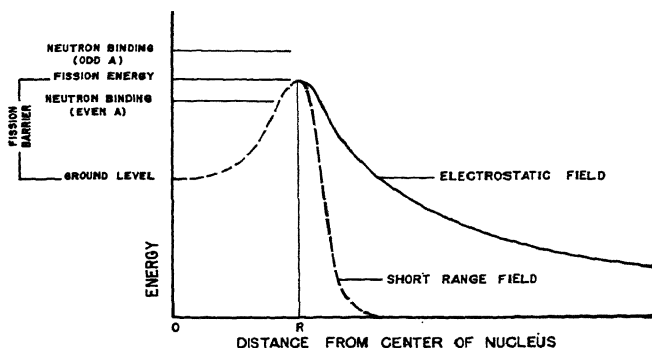


FIG. 43. Diagrammatic representation of potential fields of nucleus.

energy, gamma-ray and neutron emission are more probable than fission, but if it is greater, fission of the compound nucleus is most probable. Thus, neutrons with only thermal kinetic energies induce fission in the nuclei  $U^{235}$  and  $Pu^{239}$  with greater probability than for radiative capture since the neutron binding energies of 5.4 and 6.4 mev are greater than the respective critical energies of 5.2 and 5 mev. On the other hand, the probability of fission in  $U^{238}$  and  $Th^{232}$  becomes important only for fast neutrons since the critical energies are of the order of 1 mev greater than the neutron binding energy. In general, the fission barrier height in the heaviest nuclei with odd atomic weight is lower than the neutron binding energy, thus allowing fission to be induced by thermal neutrons.

The absolute fission cross section depends, as in all other nuclear reactions, on the neutron energy. When fission can be induced by thermal neutrons, well-defined resonance peaks may be involved at certain energies. The fission cross section at exact resonance is then many times greater than its off-resonance value. The cross section in the thermal-energy region when

resonances are absent varies as  $E^{-1/2}$ . When, because of a high fission barrier, fission can be induced only by fast neutrons, the resonance levels of the nucleus are broad and closely spaced and thus lose their identity. The cross section in this case decreases almost uniformly with increasing energy approximately as  $E^{-1}$ .

The reaction time of nuclear processes or, more correctly, the mean life of a highly excited nucleus is in general exceedingly small,  $< 10^{-10}$  sec. It is to be expected therefore that under conditions which make fission a highly probable process compared to radiation and neutron emission the mean time for fission following neutron capture should be of this order of magnitude. This was demonstrated in experiments by Feather [13], indicating that fission may occur in an interval at least as short as  $5 \times 10^{-13}$  sec, and by

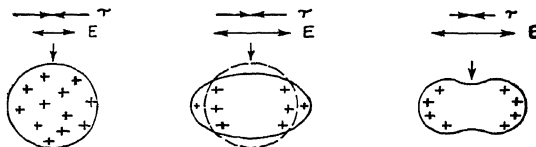


FIG. 44. Simplified concept of deformation of liquid-drop nucleus undergoing fission. The unexcited nucleus is represented as a sphere with uniform distribution of charge (protons). The relative strengths of the surface tension  $\tau$  and electrostatic forces  $E$  are indicated by lengths of vectors. Deformation of the drop induced by external forces causes a reduction in  $\tau$  at the point of least curvature and an increase in the repulsive force  $E$  due to the redistribution of charge to regions of large curvature.

Wilson [14] who showed that less than  $5 \times 10^{-5}$  of the fissions are delayed as long as  $10^{-8}$  sec.

**6.3. Stability of Heavy Nuclei.** The stability of heavy nuclei against fission depends on the relative magnitudes of the short-range nuclear forces responsible for an effective surface tension and the repulsive electrostatic field of the protons. Since the volume of the nucleus is directly proportional to the number of particles,  $A$ , it contains [7], the energy associated with the short-range forces maintaining a stable spherical nucleus is proportional to the nuclear surface,  $4\pi R^2 \sim A^{2/3}$ . Counteracting this, the uniform distribution of charge  $eZ$  gives rise to an electrostatic energy proportional to  $Z^2 A^{1/3}$  which increases more rapidly with nuclear size than does the surface tension. A critical value of the ratio of the two fields, expressed by  $Z^2(A^{1/3}/A^{2/3}) = Z^2/A$ , is attained for increasing nuclear size when the electrostatic field exceeds the nuclear binding forces and the nucleus is no longer stable against spontaneous fission. A semiempirical calculation by Bohr and Wheeler [4] leads to a critical value of  $Z^2/A = 47.8$ .

Nuclei with  $Z^2/A$  only slightly smaller ( $\sim 15$  per cent) than the critical value exhibit a marked stability against spontaneous fission when unexcited,

but division becomes highly probable for small deformations in the spherical form induced by external excitation. Classically, the minimum or critical excitation energy required to induce fission is equivalent to the height of the potential or fission barrier formed by the difference of the attractive nuclear field and repulsive electrostatic field. Alternatively, it is the energy necessary to induce a deformation of a liquid drop for which the cohesive forces become smaller than the repulsive forces within the drop. The height of the barrier decreases with increasing value of  $Z^2/A$ , and when  $Z^2/A = 47.8$ , the fission barrier vanishes entirely.

For nuclei with  $Z^2/A$  near the limiting value, the critical excitation energy for fission was calculated by Bohr and Wheeler [4] who gave the expression

$$E = 4\pi r_0^2 \tau A^{2/3} \left[ \frac{98}{135} (1-x)^3 - \frac{11,368}{34,425} (1-x)^4 + \dots \right] \quad \text{mev}$$

where  $x = Z^2/47.8A$

$$\begin{aligned} 4\pi r_0^2 \tau &= 14 && \text{mev} \\ r_0 &= 1.47 \times 10^{-13} && \text{cm [4,8]} \end{aligned}$$

When the critical energy of a nucleus is smaller than the neutron binding energy, as it is for  $\text{U}^{234}$ , fission can follow from the capture of thermal neutrons. When on the other hand it is greater than the binding energy, fission becomes probable only when the kinetic energy of the incident neutron is great enough to make up the deficit. An estimate of the neutron binding energy in heavy nuclei, therefore, is important to determine the neutron kinetic energy necessary to induce fission. For nuclei with masses greater than  $A = 230$ , the neutron binding energy has a value between 5 and 7 mev. On capture of a neutron this energy plus the neutron kinetic energy is contributed to excitation of the compound nucleus. From the relation  $Mc^2 = E$ , the binding energy  $E_b$  can be calculated from the mass difference of the initial and compound nuclei

$$E_b = 931(M_A - M_{A+1} + n)$$

for which the masses  $M_A$  and  $M_{A+1}$  may be estimated from the semiempirical relation given in Sec. 1.4.

Spontaneous fission is possible but relatively improbable in unexcited nuclei and in nuclei with less than the critical excitation energy. The half-life for spontaneous fission from the ground state appears to be  $10^{17}$  to  $10^{22}$  years for fissionable nuclei such as thorium, uranium, and plutonium.

**6.4. Fission Fragments.** The mass distribution of fission fragments exhibits a marked asymmetry about a value of one-half the mass of the initial compound nucleus. Division into two fragments of nearly equal mass occurs with little probability,  $\sim 0.01$  per cent; instead, as shown in

Fig. 45, the masses tend to group into symmetrical distributions about a light and heavy component. The most probable fragments from  $U^{235}$ , for example, are those with masses 95 and 140, corresponding to the maxima of the empirical yield curve. These values appear to be nearly the same also for  $U^{238}$  and  $Th^{232}$ . The total variation in mass ranges from approximately

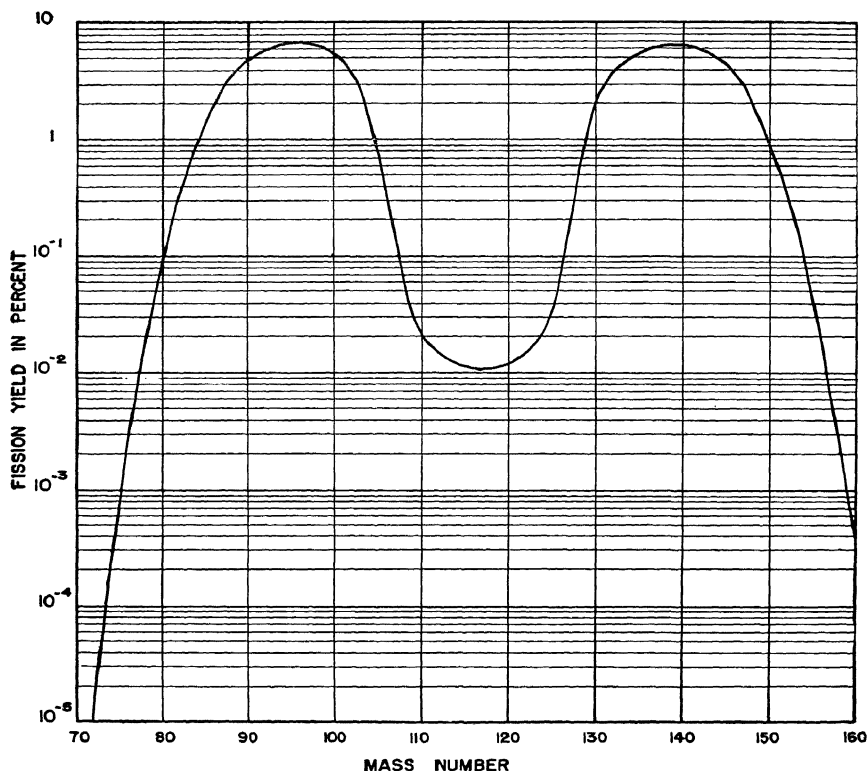


FIG. 45. Yield of fission products from  $U^{235}$  as a function of mass number. [From *J. Am. Chem. Soc.* 68, 2435 (1946).]

70 to 160 although a very high percentage of the yield is confined to the two mass groups 80 to 110 and 128 to 154. In all instances the sum of the atomic weights of the two fragments is somewhat smaller than the initial nucleus owing to the loss of two to three prompt neutrons. No satisfactory theory has as yet been provided which accounts for the asymmetry in fission yield.

The distribution in charge of primary fission fragments is less easy to establish, and conclusive experimental or theoretical results have not yet been reported. It is likely however that there is no unique charge associated with each mass but rather a number of possible values of charge distributed about a mean value for each mass.

The greatest portion of the total energy released in fission appears as kinetic energy of the fragments. A rough estimate of the recoil energy may be calculated from the electrostatic analogy of two charged particles. Assuming the fission fragments to be two spheres with charges  $Z_1$  and  $Z_2$  and initially in contact, the potential energy is then

$$E = \frac{eZ_1Z_2}{1.47 \times 10^{-13}(A_1^{1/3} + A_2^{1/3})} \quad \text{ergs}$$

where  $A_1, A_2$  = atomic weights of fragments

The recoil energy computed by this method is approximately 200 mev, about 25 per cent greater than the best experimental value. Measurements of the average recoil energy have been made by many investigators [10,15,21,22, and others] who have reported values varying from 120 to nearly 200 mev. The total kinetic energy imparted to the fragments does not have a unique value but exhibits a statistical variation about a mean value of about 160 mev [21,23,24,25,26]. The distribution appears to be symmetrical about this value and has a spread from 120 to nearly 200 mev. Each of the two fragments from a single fission share the total kinetic energy inversely as their respective masses,  $E_1/E_2 = M_2/M_1$ . When the observed energies of the fragments are plotted separately, two symmetrical distributions are obtained with mean values of approximately 60 and 92 mev, corresponding to the heavy and light components, respectively. The width of the low-energy peak at one-half its maximum value is approximately 25 mev, and for the high-energy component it is about 16 mev. The characteristics of the energy-distribution curve appear to be nearly the same for  $\text{Th}^{232}$ ,  $\text{U}^{235}$ ,  $\text{U}^{238}$ , and  $\text{Pu}^{239}$ . Although small differences for these isotopes have been found, the variation in values reported for any one fissionable isotope appears to be greater than the differences between the energy distributions for the four isotopes above.

**6.5. Absorption and Range of Fission Fragments.** The interaction of fission fragments with matter involves the same processes as in the stopping of lighter charged particles such as alpha particles, namely, ionization and elastic nuclear collisions. Nevertheless, the characteristics of the rate of energy loss and the range of fission fragments as functions of velocity are strikingly different than for alpha particles owing to their great mass, high charge number, and relatively low velocity. The range is small, the ionization is many times more intense than that produced by alpha particles, and

elastic collisions with nuclei take a relatively more important part in the stopping of fission fragments than in the stopping of lighter charged particles.

Despite the great kinetic energy (160 mev total) with which the fragments recoil at the moment of separation in the fission process their velocities are relatively low. The most probable initial velocities of the light and heavy groups are found to be about  $13.3 \times 10^8$  cm per sec and  $9.0 \times 10^8$  cm per sec, respectively [33]. These velocities are lower than the orbital velocities of electrons in the innermost shells of the fragments. Such electrons with velocities greater than that of the fragment are most likely to be retained as a permanent core from the instant the two fragments separate. Although the fragments are never completely stripped of electrons, the initial deficiency at the moment of separation has been found on the average to be about 20 electrons for the light group and about 22 for the heavy group of fragments. As a fragment is slowed down by an absorbing medium, additional electrons are captured and the charge of the particle rapidly decreases until the full complement of electrons is attained when the fragment is brought to rest.

The specific ionization along the path of the fragment does not follow the Bragg curve which describes the ionization produced by protons and alpha particles. Instead, the ionization is most intense at the beginning of the path where the fragment has its greatest charge and then diminishes rapidly along the path owing to the decrease in charge resulting from electron capture as the particle loses momentum. The fragment continues to ionize until brought to rest, but below a velocity of about  $2.5 \times 10^8$  cm per sec the electron shells remain nearly filled and energy loss by ionization is negligible.

Energy loss at low velocities,  $< 2.5 \times 10^8$  cm per sec, is due almost entirely to elastic collisions with nuclei. Large fractions of the fragments' residual kinetic energy and momentum may be lost in single collisions, and relatively few collisions are necessary to stop the fragment. The average number of collisions depends on the atomic weight of the absorbing medium, but even in light elements it is probably less than ten. The recoil nuclei are observed in cloud chambers as short branches of varying length extending from the track of the fission fragment. In light gases such as hydrogen the fragment track remains relatively straight and shows many branches, whereas in gases of medium atomic weight such as xenon, collisions are fewer and the fragment track is more strongly deflected.

The stopping of fission fragments in the first part of the range where ionization is most important results in a nearly linear relation between velocity and range, as shown in Fig. 46. In the second part, below a velocity of  $2.5 \times 10^8$  cm per sec, the range is influenced solely by elastic collisions and decreases rapidly to zero. The great variation in the number of collisions that fragments undergo strongly influences the shape of the second part of the velocity-range curve and makes straggling large. The straggling is



further enhanced by the variation in initial energy, charge, and mass of the fragments. Efforts to calculate a stopping formula and range-energy relation have been unsuccessful, mainly because the charge of the fragment is a rapidly changing and, as yet, unknown function of velocity. The stopping theory proposed by Bohr [25] is, however, confirmed in principle by results of range and stopping-power measurements [33,34,35]. As is to be expected from the mass and energy division of fission fragments, short- and long-range groups are found which have been shown to correspond to the light and heavy fragments, respectively. Ranges of the two groups in various gases are given

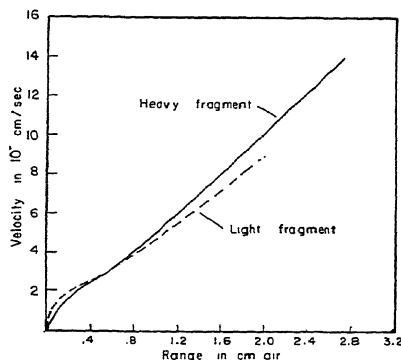


FIG. 46. Velocity-range curves of light and heavy fission fragments in air [34].

in Table 21. Table 22 gives the stopping power of various solid absorbers determined by Segre and Wiegand (37) from absorption measurements with thin foils. More detailed ranges are given in Table 23.

TABLE 21. RANGES OF FISSION FRAGMENTS IN GASES [35]

Gas	Short-range group, mm	Long-range group mm
Hydrogen.....	17.7	21.1
Deuterium.....	18.9	22.5
Helium.....	23	28
Argon.....	19.4	23.9
Xenon.....	18	23

**6.6. Radioactive Chains.** The excitation energy of fission fragments which still remains after evaporation of prompt neutrons is subsequently lost by emission of delayed neutrons, beta particles, neutrinos, and gamma rays. The remaining excess of neutrons is transformed to protons by successive beta decay or, less frequently, lost by delayed neutron emission, until the

A few of the many possible initial fission fragments are known to be stable, whereas others decay by the emission of as many as six successive beta particles. On the average each fragment undergoes three beta transformations accompanied by its characteristic isomeric transitions. Many of the

TABLE 23. RANGES OF FISSION FRAGMENTS [36]

Extrapolated and mean ranges of plutonium fission fragments in normal air, and the straggling as indicated by the widths at half-height of the differential range curves.

Mass number	Isotope	Normalized extrapolated range, cm	Normalized mean range, cm	Average width at half-maximum, per cent
83	2.4-h Br	2.895	2.65	13.4
91	9.7-h Sr	2.738	2.55	11.4
92	3.5-h Y	2.717	2.55	10.5
93	10-h Y	2.697	2.53	10.1
(94)	20-m Y	2.687	2.52	10.5
97	17-h Zr	2.661	2.50	10.7
99	67-h Mo	2.635	2.48	10.8
105	36.5-h Rh	2.587	2.42	11.4
109	13.4-h Pd	2.508	2.35	10.7
112	21-h Pd	2.416	2.24	13.4
117	1.95-h In	2.246	2.08	10.1
127	93-h Sb	2.248	2.09	11.9
129	4.2-h Sb	2.243	2.09	12.5
132	77-h Te	2.198	2.05	11.5
133	60-m Te	2.180	2.04	11.8
(134)	43-m Te	2.180	2.04	11.4
140	12.8-d Ba	2.080	1.92	12.6
143	33-h Ce	2.040	1.89	11.8
149	47-h Bi	1.977	1.82	13.1
(157)	15.4-h Eu	1.949	1.79	15.1

end activities of the observed chains correspond to earlier known activities produced by other processes.

The total decay energy associated with the chain activities of the two fragments has been measured by several investigators but with conflicting results because of the difficulties encountered in evaluating with accuracy the energy associated with the very short half-lives and penetrating gamma rays. The average total decay energy is probably between 20 and 30 mev per fission. The fraction of the total energy contributed separately to beta particles and gamma rays is not certain, but Way and Wigner [16] have given an empirical formula for the rate of energy loss by gamma rays in the interval  $10 < t < 10^7$  sec as

$$\bar{E}_\gamma = 1.26t^{-1.2} \quad \text{mev (fiss)/sec}$$

and for the average total energy

$$\bar{E}_t = 2.66t^{-1.2} \quad \text{mev (fiss)/sec}$$

Experiments by Bernstein *et al.* [17] on the yield of gamma rays with

TABLE 24. YIELD OF HARD GAMMA RAYS FROM FISSION PRODUCTS [17]

Half-life	Energy, mev	Gamma rays per fission
2.5 s	3.4	0.00015
41 s	2.25	0.0042
2.4 m	2.65	0.045
7.7 m	3.0	0.038
27 m	2.6	0.046
1.65 h	2.62	0.132
4.4 h	(3)	1.58
53 h	(3)	0.675
Total	....	2.5

energies greater than 2.17 mev from fission products indicate about eight gamma-ray components, as shown in Table 24.

**6.7. Prompt Neutrons.** On the basis of the existence rules for stable isotopes (Sec. 1.1) it is apparent that the two fragments of medium atomic weight formed in the fission of a heavy element must contain a considerable excess of neutrons compared with stable nuclei with the same number of protons. Some of the excess neutrons therefore are very weakly bound, and in view of the high excitation energy given fission fragments in the process of division, a portion of these neutrons boil off instantly. The average number of prompt neutrons per fission has been extensively investigated, and the values that have been reported vary from 1.5 to as many as 6. The most reliable of these measurements gives values between two and three prompt neutrons per fission, on the average. Although the reported experimental results do not appear conclusive for accuracies greater than this, they tend to indicate an average value of about 2.6. This value can be regarded only as a tentative estimate until experiments demonstrate conclusively the magnitude of the second significant figure.

The observed energy distribution of prompt neutrons is found to be not only continuous but also to be very nearly a Maxwellian distribution when the translational component of the fission fragment is subtracted. The superposition of the translational component of the fragment and the Maxwellian distribution bears out the contention that the neutrons are emitted from the fragments and not in the primary fission process. The form of the distribution is consistent with the assumption that at high excitation energies many nuclear levels may be affected and because of their great density they present an essentially continuous spectrum of possible transitions by neutron emission. The most likely value of the mean energy per prompt neutron appears to be approximately 2 mev, and therefore, the total average neutron energy is 4 to 6 mev per fission.

The directional distribution of prompt neutrons has been shown by Wilson [28] to be consistent with the assumption of isotropic evaporation from the moving fission fragments.

**6.8. Delayed Neutrons.** Delayed neutrons are emitted from fission products during the course of radioactive decay. They are emitted, however, from only a few of the many possible fission products that may be formed, and consequently are low in intensity compared with prompt neutrons. The observed intensity under equilibrium conditions is  $1.0 \pm 0.2$  per cent of the abundance of prompt neutrons [18].

A delayed neutron is emitted only after the initial fission fragment has undergone beta decay. If, after beta emission, the residual nucleus still possesses an excitation energy greater than the neutron binding energy, it may decay by further beta transitions or by neutron emission; the latter process usually reduces the nucleus to the ground state. Although the neutron is emitted in an exceedingly short time after the nucleus is reduced to the appropriate excited level, the observed half-time for neutron emission following the primary fission process is controlled by the preceding beta transition and, therefore, has the same half-life. Six such half-life periods have been found for delayed neutrons as shown in Table 25.

TABLE 25. DELAYED NEUTRONS [29]

Half-life, sec	Energy, mev	Yield,* per cent	Primary fission fragment
0.05	.....	0.025	
0.43	0.420	0.085	
1.52	0.620	0.241	
4.51	0.430	0.213	
22.	0.560	0.166	$\text{I}^{137}$ [18]
55.6	0.250	0.025	$\text{Br}^{87}$ [18]

\* Relative to total neutron emission.

These six periods are observed in the fission of both  $\text{U}^{235}$  and  $\text{Pu}^{239}$  and presumably are associated with the same fission fragments from either element.

**6.9. Alpha Particles.** Alpha particles have been found to be associated occasionally with fission of both  $\text{U}^{235}$  and  $\text{Pu}^{239}$ . Farwell *et al.* [11] observed a continuous energy distribution with a maximum energy of about 16 mev and were able to show that the alpha particles were emitted within  $5 \times 10^{-6}$  sec after the primary fission process. Wollan *et al.* [20], using photographic plates, found the maximum energy to be 22 mev (40 cm range in air at normal temperature and pressure) and demonstrated that alpha particles were emitted in the primary fission process and not from the fragments. It was

TABLE 26. FISSION PRODUCT YIELDS

The following table is based on data reported by the Plutonium Project in *J. Am. Chem. Soc.*, 2411-2422 (1946). Only the initial fission fragment and the final product of each of the known chains are listed here, together with the probable yield. For a complete description of the chains, reference to the above report is suggested.

Probable fission fragment			Fission yield, per cent	Probable fission fragment			Stable product			Fission yield, per cent
Z	El.	A		Z	El.	A	Z	El.	A	
				48	Cd	115	49	In	115	0.0008
30	Zn	72					48	Cd	116	
30	Zn	73	$1.5 \times 10^{-5}$	48	Cd	117	50	Sn	117	0.01
			$10 \times 10^{-4}$	48	Cd	..	48	Cd	..	
32	Ge	75					50	Sn	118	
							50	Sn	119	
32	Ge	77	0.0091				40	Sn	120	
32	Ge	78	0.02	50	Sn	121	51	Sb	121	0.014
33	As	78		50	Sn	{ 121	51	Sb	{ 121	0.0012
33	As	81	0.125			123			123	
							50	Sn	122	
34	Se	79		50	Sn	123	51	Sb	123	0.0044
							50	Sn	124	
34	Se	83		50	Sn	125	52	Te	125	0.023
34	Se	84	0.65	50	Sn	125	?			
35	Br	82	$2.8 \times 10^{-5}$	50	Sn	126	52	Re	126	0.1
35	Br	85		51	Sb	127	53	I	127	
							52	Te	128	
35	Br	87	0.026	51	Sb	129	54	Xe	129	
35	Br	87					52	Te	130	
36	Kr	88		51	Sb	132	54	Xe	132	3.6
36	Kr	89	4.6	51	Sb	133	55	Cs	133	4.5
36	Kr	90		51	Sb	134	54	Xe	134	5.7
36	Kr	91	9.5	52	Te	131	54	Xe	131	2.8
36	Kr	94	5	52	Te	135	56	Ba	135	5.9
36	Kr	97		53	I	136	56	Ba	136	0.01
				53	I	137	54	Xe	136	
37	Rb	86	$2 \times 10^{-5}$	53	I	137	56	Ba	137	
37	Rb	92	5.1	54	Xe	138	56	Ba	138	
37	Rb	..		54	Xe	139	57	La	139	6.3
36	Kr	93		54	Xe	140	58	Ce	140	6.1
39	Y	95	6.4	54	Xe	141	59	Pr	141	5.7
42	Mo	99	6.2	54	Xe	143	60	Nd	143	5.4
				54	Xe	144	60	Nd	144	5.3
42	Mo	101		54	Xe	145	60	Nd	145	
42	Mo	102		55	Cs	142	58	Ce	142	
42	Mo	105	0.9	58	Ce	146	60	Nd	146	
43	Tc	107		60	Nd	147	62	Sm	147	2.6
							60	Nd	148	
44	Ru	103	3.7	60	Nd	149	62	Sm	149	1.4
							60	Nd	150	
44	Ru	106	0.5	60	Nd	151	63	Eu	151	
45	Rh	109	0.028			153	62	Sm	152	
				61			63	Eu	153	0.15
46	Pd	111	0.018			156	62	Sm	154	
46	Pd	112	0.011	61			64	Gd	156	0.013
				62	Sm	155	64	Gd	155	0.03
				63	Eu	157	64	Gd	157	0.0074
				63	Eu	158	64	Gd	158	0.002

also observed in these experiments that the distribution of alpha-particle tracks tended to bunch about a direction at right angles to that taken by the fragments. The frequency of occurrence of alpha particles appears to be about 1 in 250 fissions of  $\text{U}^{235}$  and 1 in 500 fissions of  $\text{Pu}^{239}$  [11].

**6.10. Fission Induced by Other Radiations.** Neutrons are the most efficient means for inducing fission in heavy elements since they are unaffected by the high electrostatic potential barrier, but charged particles and gamma rays will also induce fission when given sufficient energy. Photofission has been investigated with high-energy gamma rays, and the cross sections for uranium and thorium are reported as  $\sigma_{\text{U}} = 0.0035$  barn and  $\sigma_{\text{Th}} = 0.0017$  barn, respectively [30]. Alpha particles and deuterons are also known to induce fission, but in the case of deuterons it is not yet certain if the entire particle enters the nucleus or is first stripped of its proton so that only the neutron is captured.

**6.11. Fission of Elements below Thorium.** Despite the increasing height of the fission barrier in elements with atomic numbers lower than thorium, it was anticipated that fission could be induced in such elements with particles of very high energy. This has been verified by Perlman *et al.* [31,32] for the elements bismuth, lead, thallium, platinum, and tantalum by bombarding targets of these elements with 400-mev alpha particles, 200-mev protons, and 100-mev neutrons produced by the 184-in. Berkeley cyclotron. Bismuth, the most carefully investigated element, exhibits marked differences from fission of uranium by low-energy neutrons. Unlike uranium fission the masses of the bismuth fission products are distributed symmetrically in a single peak about mass 98-99. Also a greater proportion of the products are either stable or  $\beta^+$  active.

A qualitative explanation of bismuth fission is given [32] which suggests that only after about 12 neutrons have boiled off the compound nucleus ( $\text{Bi} + d$ ) does fission become probable; the fissionability parameter  $Z^2/A$  is then about equal to that of  $\text{U}^{236}$ . Further, if it is assumed that the fragments retain the same neutron-to-proton ratio as the light bismuth nucleus ( $n/p = 1.36$ ) then, as has been observed, the fragments with masses less than  $\sim 100$  carry off an excess number of neutrons and are  $\beta^-$  emitters, those of intermediate masses are stable, and the heavier fragments are neutron deficient and therefore  $\beta^+$  or K capture active.

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## CHAPTER 7

### RADIOACTIVITY

**7.1. Summary of Radioactivity.** Radioactivity refers only to those processes by which unstable nuclei decay by loss of their excitation energy to form known stable nuclear species. These processes are distinct from the primary processes of nuclear interaction in which unstable or excited nuclei are formed. Radioactive decay proceeds at a rate proportional only to the number of unstable nuclei present and is wholly independent of external influences, whereas the primary process of formation depends upon the type and energy of the bombarding radiation. In addition, the primary process of formation of an excited nucleus takes place in a time of the order of  $10^{-10}$  sec or less, while the probable time for decay of an unstable nucleus is enormously longer and varies over the range from  $10^{-7}$  sec (RaD) to  $4.4 \times 10^{17}$  sec (thorium).

All known unstable nuclei decay only by emission of one or more of the following radiations: negatron, positron, neutrino, gamma ray, alpha particle, and neutron. In general, decay proceeds by several discrete energy steps or transitions to lower nuclear quantum levels until the ground state of a stable nuclear species is reached. In most cases this involves the emission of a charged particle and one or more gamma rays. In many instances, particularly in medium and heavy nuclei, decay may occur by one of several alternative processes involving different sets of quantum levels and sometimes different radiations. The probabilities for the alternative processes is given directly by the observed fractions of different radiations when the decay scheme is known.

A summary of the processes that are known to occur in radioactive decay is given below. More detailed discussions will be found in those sections describing the specific radiations.

*a. Negatron Emission (Beta Decay).* A beta particle and neutrino are emitted simultaneously and share in any proportion the total fixed energy corresponding to a discrete level transition in the nucleus. The observed beta kinetic energy varies from zero to a well-defined maximum corresponding to the total transition energy. The residual or daughter nucleus is greater in charge by one electronic unit, and the exact atomic weight is diminished by only the mass equivalence of the maximum kinetic energy of the beta particle emitted. The statistics of the nucleus remain unchanged, but the nuclear spin is altered by an integral multiple of  $\hbar/2\pi$ .

*b. Positron Emission (Beta Decay).* A positron and a neutrino are emitted simultaneously and share in any proportion the total discrete energy of the level transition. The observed positron kinetic energy may take any value from zero up to the well-defined total transition energy. The residual atom is smaller in charge by one electronic unit, and its exact atomic weight is diminished by an amount equal to the mass equivalence of the maximum kinetic energy plus twice the electron rest mass. The statistics of the nucleus remain unchanged, but the spin is altered by an integral multiple of  $\hbar/2\pi$ .

*c. K Capture.* An alternative process to positron decay, K capture is often found to compete with positron emission for the same level transition in the transformation proton  $\rightarrow$  neutron. Unlike positron decay, however, a monoenergetic neutrino is emitted and K x-radiation characteristic of the daughter substance is emitted. The residual atom is smaller in charge by one electronic unit, and its exact atomic weight is diminished by the mass equivalence of the kinetic energy carried off by the neutrino. The statistics of the nucleus remain the same, but the spin is altered by an integral multiple of  $\hbar/2\pi$ .

*d. Isomeric Transition (Gamma Emission).* Gamma rays are always monoenergetic for any one nuclear level transition, but several gamma rays of different energy  $h\nu$  may be emitted in cascade in the decay of a single atom. The charge of the residual nucleus remains unaltered, the spin is altered by an integral multiple of  $\hbar/2\pi$ , and the exact atomic weight is reduced by the amount  $h\nu/c^2$ .

*e. Internal Conversion.* Although internal conversion is not a nuclear phenomenon in the decay process, it is frequently observed to accompany gamma emission. Internal conversion (I. C.) is essentially the photoelectric absorption of a gamma ray by an orbital electron of the atom from whose nucleus the gamma ray is emitted. K electrons are often observed to have the highest probability for emission, L electrons next, etc. The electron is ejected with a kinetic energy equal to the gamma-ray energy  $h\nu$ , minus the binding energy (ionization potential) of the electron in the atom. Ejection of conversion electrons is accompanied by characteristic K, L, M, and possibly N, x-radiation. The probability for the process is given in terms of the conversion coefficient  $\alpha$  which is numerically equal to the ratio of the number of conversion electrons ejected to the number of gamma rays emitted:

$$\alpha = \frac{N_K + N_L + \dots}{N_\gamma}$$

where  $N_\gamma$ ,  $N_K$ , . . . are the observed numbers of gamma rays and K, L, . . . electrons.

*f. Alpha Decay.* With a single exception (lithium) alpha-particle emission is always monoenergetic. Although only one alpha particle is emitted per disintegration, many energy groups may be observed corresponding to different and more or less probable level transitions in the same species of nucleus. The residual atom is smaller in charge by two electronic units and is diminished in mass by an amount approximately equal to the rest mass of the alpha particle plus the kinetic energies of the particle and recoil nucleus, plus the rest mass of two electrons.

**7.2. Law of Radioactive Decay.** Radioactive decay is a statistical process following well-established rules. A single radioactive nucleus possesses a fixed probability of disintegration per unit time which is characteristic of the particular isotope and its state of excitation. Aside from these two factors, the probability of decay, and hence the rate of decay of a macroscopic quantity of the radioisotope, is wholly independent of external influences such as temperature, pressure, chemical reagents, and the means by which it is produced. Since each disintegration is a statistically independent event, the average number of nuclei which disintegrate per unit time is proportional to the number of nuclei of the particular species in a prescribed state of excitation that are present at any instant.

$$\frac{dN}{dt} = -\lambda N$$

where  $N$  = number of nuclei present at time  $t$

$\lambda$  = disintegration constant; factor of proportionality characteristic of the isotope and its state of excitation

In practice the quantity  $N$  frequently is given any convenient dimension and may be referred to as the number of nuclei, activity, mass, or some wholly arbitrary unit convenient to the particular measuring device.

**7.3. Fluctuations.** The actual number of disintegrations observed per unit time fluctuates about a mean value  $N$  as a consequence of the random character of the disintegration process. The probability of observing  $M$  disintegrations per unit time when the average value is  $N$  is given by the Poisson probability distribution formula:

$$P_M = \frac{N^M}{M!} e^{-N}$$

When the average value is large and the difference  $N - M$  is small, the probability distribution may be represented approximately by Gauss' formula:

$$P_M = \frac{1}{\sqrt{2\pi N}} e^{-\frac{(M-N)^2}{2N}}$$

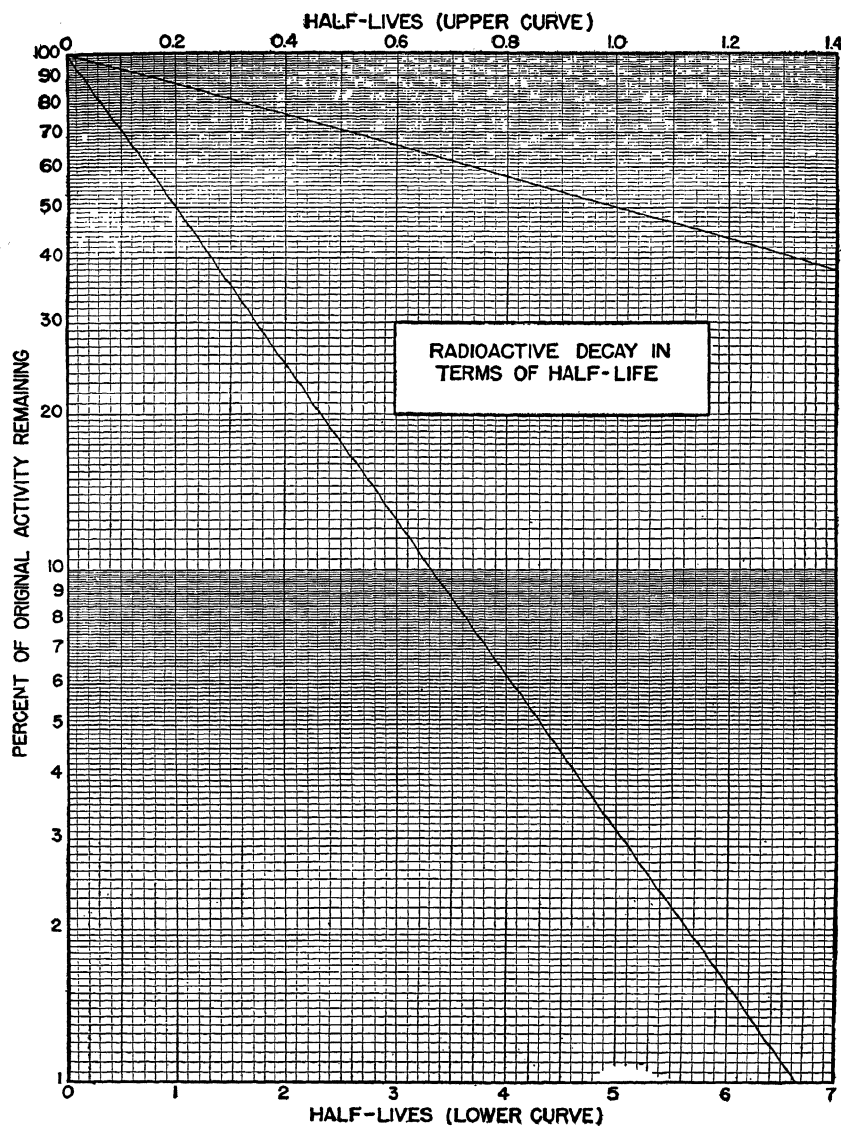


FIG. 47.

An estimate of the statistical error in the measurement of an activity  $N$  is expressed usually in terms of the absolute probable error given by

$$r = 0.6745 \sqrt{N}$$

or in terms of the per cent probable error by

$$r\% = 67.45 \frac{1}{\sqrt{N}}$$

It is assumed, of course, that the period of observation is very small compared to  $1/\lambda$ .

**7.4. Simple Decay.** The observed decay of a given quantity of radioactive isotope follows a simple exponential law as is readily apparent on integrating the expression  $dN/dt = -\lambda N$  in Sec. 7.2. This assumes, of course, that the isotope is not at the same time being produced by some external agent. If  $N_0$  is the initial number of atoms of the radioactive isotope, the number of atoms remaining after a time  $t$  is

$$N = N_0 e^{-\lambda t}$$

where  $\lambda$  = decay constant in units of reciprocal time

However, the decay of an isotope is usually expressed in terms of half-life  $T$ , which is the time required for an initial number of radioactive nuclei to be reduced by one-half. The number of atoms present at time  $t$  is then

$$N = N_0 e^{-0.693t/T}$$

Also frequently used is the mean life  $\tau = T/0.693 = 1/\lambda$ , which is the time required for an initial quantity of radioisotope to be reduced by  $1/e$ . The activity of any substance as a function of the number of half-lives is plotted in Fig. 47 for convenient reference.

**7.5. Growth of Radioactivity.** When a radioisotope is produced at a constant rate, for example, in a cyclotron or as a daughter substance of

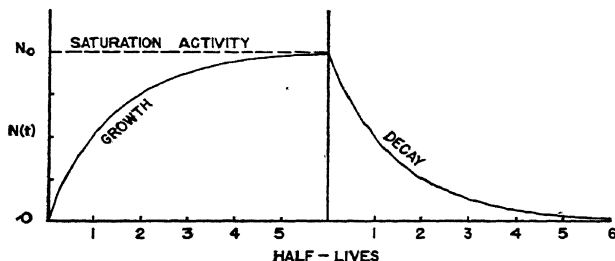


FIG. 48. Growth of a radioactive isotope that is produced at a constant rate, e.g., in a cyclotron, and decay after production is stopped.

uranium, the number of atoms of the product radioisotope after a time  $t$  is

$$N = N_o(1 - e^{-0.693t/T})$$

where  $N_o$  = saturation activity; the activity at which the rate of production of the isotope equals rate of its decay. This is equivalent to the rate of formation divided by  $\lambda$

**7.6. Radioactive Substance Produced by Parent of Long Half-life.** When a radioactive daughter substance is produced for which the half-life  $T_2$  is short compared with that of the radioactive parent, *i.e.*,  $T_1 \gg T_2$ , and assuming that only the parent substance is present initially, the quantity of the daughter substance after time  $t$  is

$$N_2 = N_o \frac{T_2}{T_1} (1 - e^{-0.693t/T_1})$$

where  $N_o$  = initial quantity of parent substance

After many half-lives of the daughter, the parent and daughter are in secular equilibrium and the decay of the latter is governed now by the decay of the parent. The quantity of the daughter is then proportional to the ratio of the half-lives of the two substances:

$$N_2 = N_o \frac{T_2}{T_1}$$

**7.7. Parent and Daughter Substances of Comparable Half-lives.** When the half-lives of parent and daughter are of the same order of magnitude and only the parent is present initially, the activity of the daughter substance at

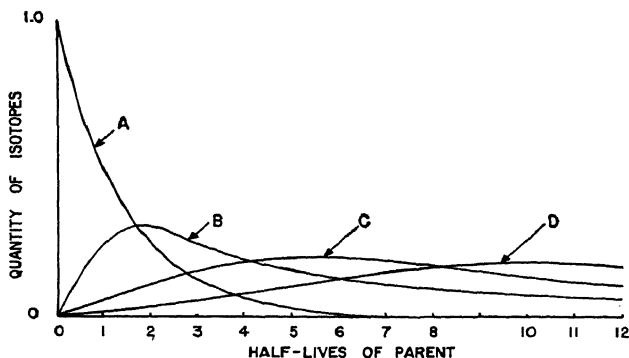


FIG. 49. Growth and decay of a series of daughter substances in a radioactive chain  $A \rightarrow B \rightarrow C \rightarrow D \rightarrow \dots$ . Only the substance  $A$  is present initially.

any later time is given by

$$N_2 = \frac{T_2}{T_1 - T_2} N_0 (e^{-0.693t/T_1} - e^{-0.693t/T_2})$$

The activity of the daughter increases to a maximum and then decays after a sufficiently long time at a rate corresponding to the longer of the two half-lives. The ratio of daughter to parent substance in transient equilibrium is

$$N_2 = \frac{T_2}{T_2 - T_1} N_0$$

This mode of radioactive decay is shown in Fig. 49. The area under all curves must be equal since the same number of atoms is involved in the complete decay of parent and daughters.

**7.8. Decay of  $n$ th Component in a Radioactive Chain.** If a given initial quantity  $N_0$  of radioisotope decays into a series of radioactive daughter substances, the quantity of the  $n$ th successive daughter substance after a time  $t$  is given by [1]

$$N = N_0(a_1 e^{-0.693t/T_1} + a_2 e^{-0.693t/T_2} + \dots + a_n e^{-0.693t/T_n})$$

where

$$\begin{aligned} a_1 &= \frac{T_1^{n-1}}{(T_1 - T_2)(T_1 - T_3) \cdots (T_1 - T_n)} \\ a_2 &= \frac{T_2^{n-1}}{(T_2 - T_1)(T_2 - T_3) \cdots (T_2 - T_n)} \\ a_n &= \frac{T_n^{n-1}}{(T_n - T_1)(T_n - T_2) \cdots (T_n - T_{n-1})} \end{aligned}$$

$T_1, T_2, \dots, T_n$  = half-lives of parent and successive daughter substances

**7.9. Radioactivity Units.** *a. Curie.* 1 curie = 0.66 mm<sup>3</sup> of radon at 0°C, 760 mm Hg.

Definition: A curie is the quantity of radon in radioactive equilibrium with 1 gm of radium.

The evaluation of the curie depends on the determination of the absolute disintegration rate of radium since, from its definition, the curie is that quantity of radon which disintegrates at the same rate as radium. Measurements of the decay rate have in the past differed in magnitude by as much as 10 per cent, and in order to establish some standard an arbitrary value of  $3.70 \times 10^{10}$  disintegrations per second per gram of radium was recommended by the International Radium Standard Commission until agreement can be reached on the third significant figure. Initially, the curie applied only to radon in equilibrium with radium; however, its extension to the other decay

products in equilibrium with radium was recommended by the Commission in 1930 [2]. A curie of any radium product, therefore, is that quantity of the isotope which decays at the rate of  $3.7 \times 10^{10}$  disintegrations per sec.

In addition to the restricted use of the curie recommended by the Commission, it has become widely adopted as a measure of the quantity of any radioactive isotope. More generally, the fractional units millicurie (mc) and microcurie ( $\mu$ c) are used; these are the quantities of the isotope that decay at the rates of  $3.7 \times 10^7$  and  $3.7 \times 10^4$  disintegrations per sec, respectively. The weight or the number of atoms of an isotope equivalent to 1 millicurie is directly proportional to the half-life

$$N = 3.7 \times 10^7 \frac{T}{0.693}$$

$$W = 3.7 \times 10^7 \frac{AT}{N_0}$$

where  $N$  = number of atoms per millicurie of isotope

$W$  = weight in gm per millicurie of isotope

$T$  = half-life of isotope, in sec

$A$  = atomic number

$N_0$  = Avogadro's number

Failure to distinguish between total ionizing events and total disintegrations has led to occasional confusion in the use of the curie for some isotopes that do not have simple decay schemes. When beta particles and gamma rays are emitted, the number of events detected (corrected for geometry, absorption, and efficiency) is sometimes not equal to the number of disintegrations, and it is essential, therefore, to know the decay scheme for the radioisotope. This must include the number of particles per disintegration, the percentage and energy of each, and the percentage of internal conversion of gamma rays if they are present. Only when this detailed information is known can the disintegration rate of an isotope be ascertained from measurements of its radiation and its quantity expressed in curies.

Detection of the beta particles, for example, from a radioisotope that emits a beta particle and gamma ray in succession may lead to a disintegration rate that is too high if there are also internal conversion electrons. For the same reason, detection of the gamma rays may well lead to a low value if the conversion coefficient is unknown. Similarly, the decay rate of a radioisotope that disintegrates by either positron emission or K capture cannot be determined unless the relative probabilities of the two processes are known, or both the positrons and x-rays are detected.

*b. Rutherford.* 1 rutherford =  $1/(3.7 \times 10^4)$  curie =  $\frac{1}{37}$  millicurie.



Definition: A rutherford is the quantity of any radioisotope that decays at the rate of  $10^6$  disintegrations per second.

The rutherford has been proposed as a new unit of quantity for all radioisotopes with the exception of the decay products of radium for which the curie applies [3]. Its value was arbitrarily chosen so that 1 rutherford is in the order of magnitude of quantities used in tracer and therapeutic applications.

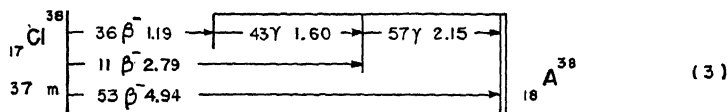
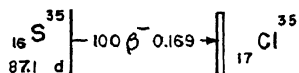
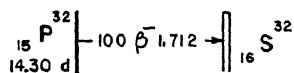
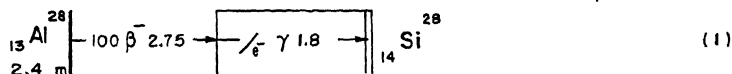
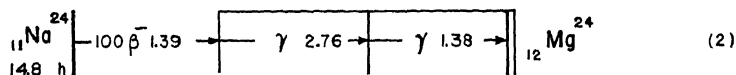
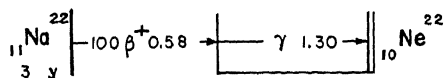
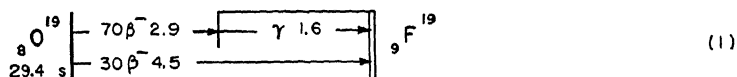
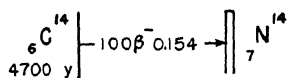
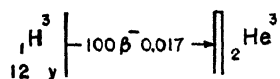
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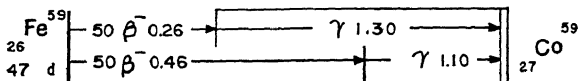
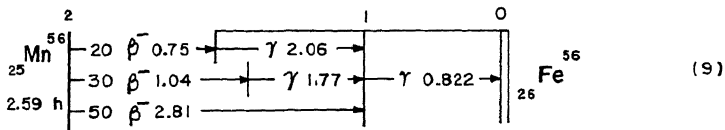
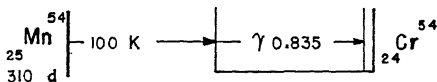
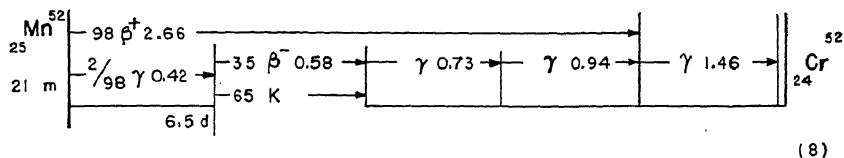
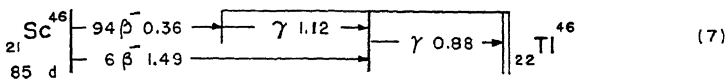
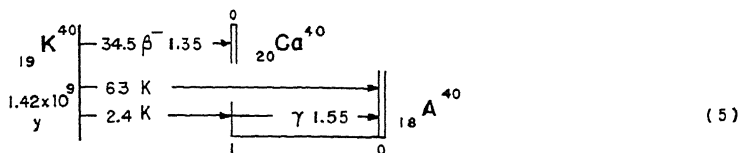
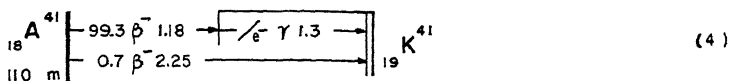
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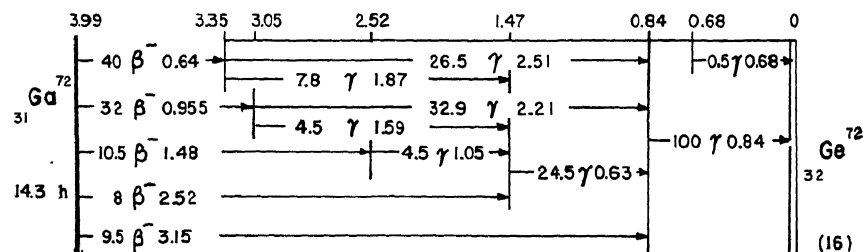
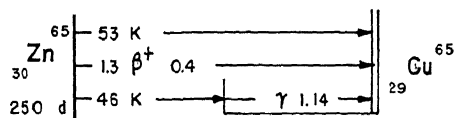
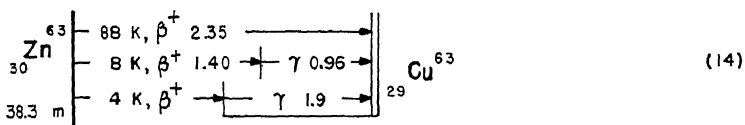
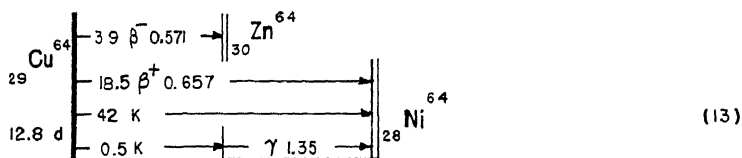
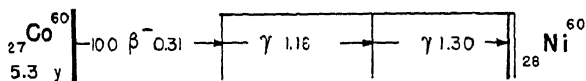
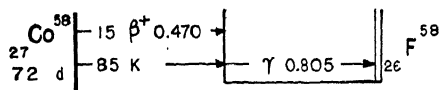
**7.10. Decay Schemes.** The decay schemes given below are intended to convey as much essential information as possible about the radiations associated with radioactive decay of certain isotopes. It must be recognized that the experimental data are not yet complete nor are they always conclusive. Many of the decay schemes therefore are still tentative, and most are incomplete so far as detailed information, such as conversion coefficients, is concerned.

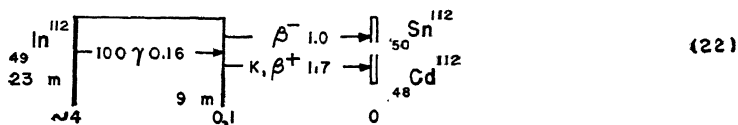
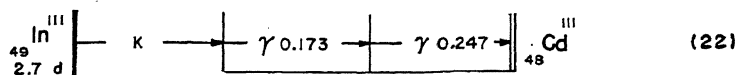
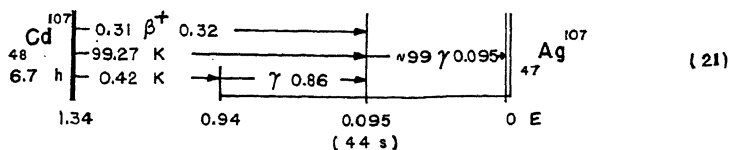
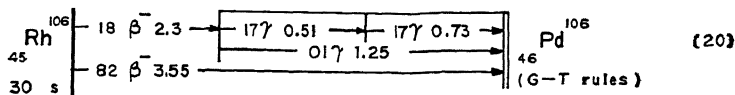
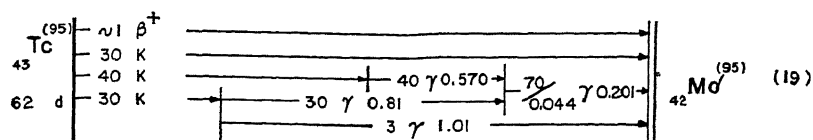
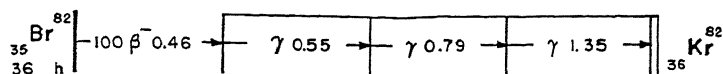
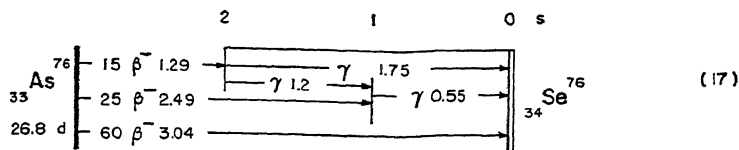
The transitions from the unstable states of a nucleus to its ground state proceed from left to right. The first transition is indicated, for example, by 70  $\beta^-$  2.9. The first number is the percentage of disintegrations in which the particular transition ( $\beta^-$  emission) occurs, followed by the type of radiation and finally, the energy in mev, except in K capture. Isomeric transitions are indicated in the same manner except that when the internal conversion coefficient is known it is given as the denominator of a fraction before the Greek letter  $\gamma$ , the numerator, when given, indicating the percentage of disintegrations in which the particular transition occurs. The conversion coefficient given here is defined by  $f = N_e/(N_e + N_\gamma)$ , where  $N_e$  and  $N_\gamma$  are the numbers of conversion electrons and gamma rays associated with the transition which are emitted per transition. When the spin and energy of particular levels are known, these are indicated above or below the vertical bar indicating the level. The ground state of the stable nucleus is indicated by a vertical double bar and designated by the  $Z$ , symbol, and  $A$  of the stable nucleus. Excited levels of the same nucleus are indicated by a horizontal line connecting the indicated levels.

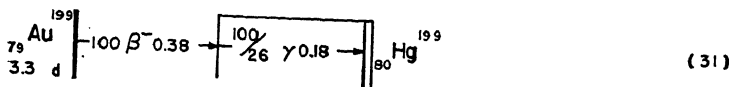
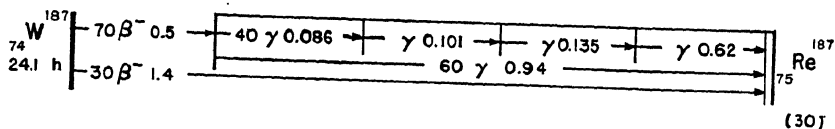
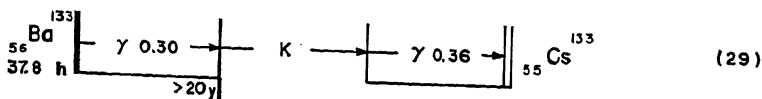
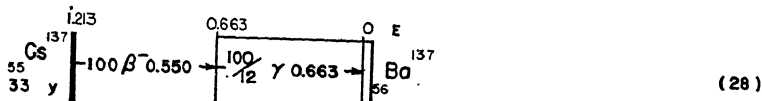
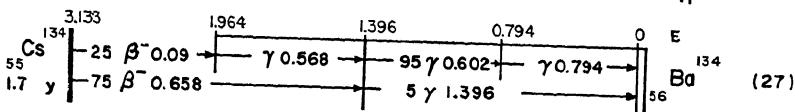
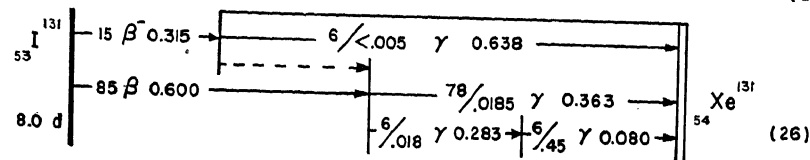
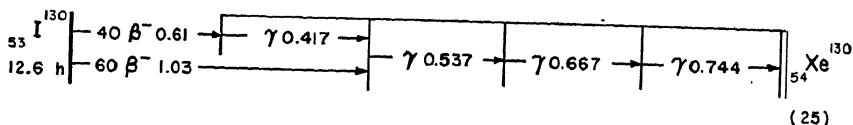
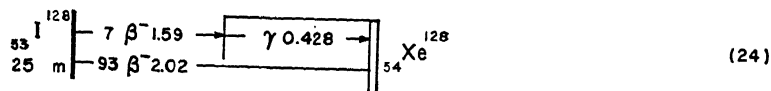
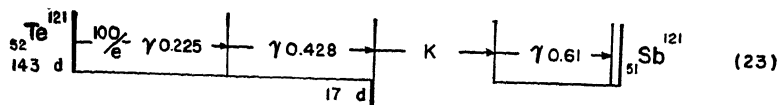
The numbers in parentheses in the schemes refer to the numbered references given on page 177.











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**7.11. The Natural Radioactive Series.**

The three radioactive chains (see Figs. 50, 51, and 52) growing out of the long-lived parent isotopes  $\text{Th}^{232}$ ,  $\text{AcU}^{238}$ , and  $\text{UI}^{238}$ , contain those radioactive isotopes that, together with  $\text{K}^{40}$  and  $\text{Rb}^{87}$ , comprised the only radioactive isotopes known before the discovery in 1932 of artificially induced radioactivity. The members of these series are found to occur in natural sources and in this sense are actually the only "natural" radioactive series. In recent years, however, numerous short side branches have been added to these series as a result of the discovery of many new isotopes produced artificially. All of these branches consist of isotopes with relatively short half-lives and therefore do not occur naturally. The longest of these branches, as shown in Fig. 50, contains five members, and, although it runs into the uranium-radium series, it has been separately named the protactinium series after  $\text{Pa}^{230}$ , its longest-lived member.

A new series, completely independent of the three naturally occurring series, has also been found whose leading members were produced artificially. Although the series starts with  $\text{Pu}^{241}$ , it has been named the neptunium series after its longest-lived member  $\text{Np}^{237}$ . This chain would appear to complete

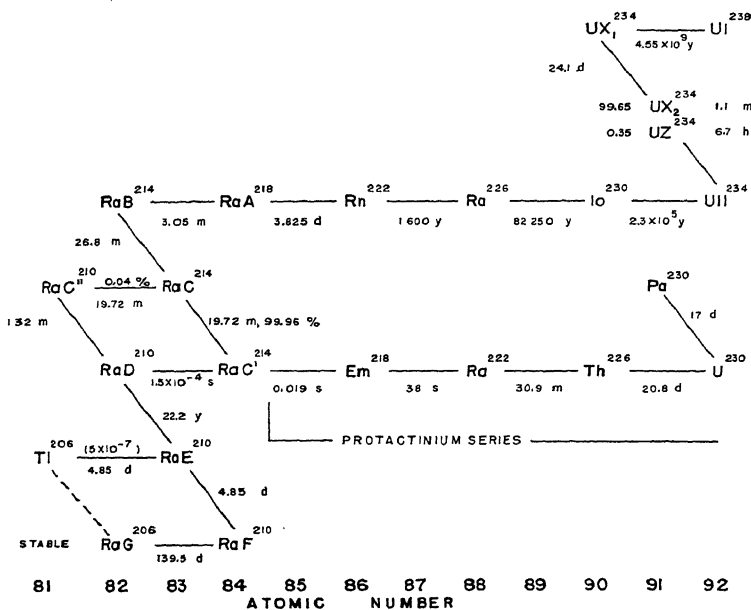


FIG. 50. Uranium-radium series and protactinium series.

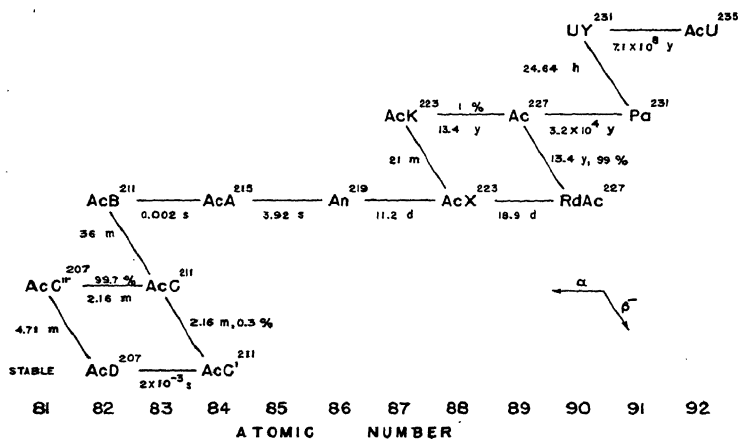


FIG. 51. Actinium-uranium series.



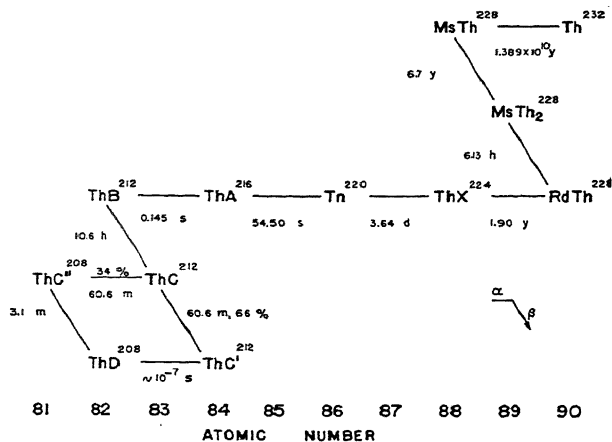


FIG. 52. Thorium series.

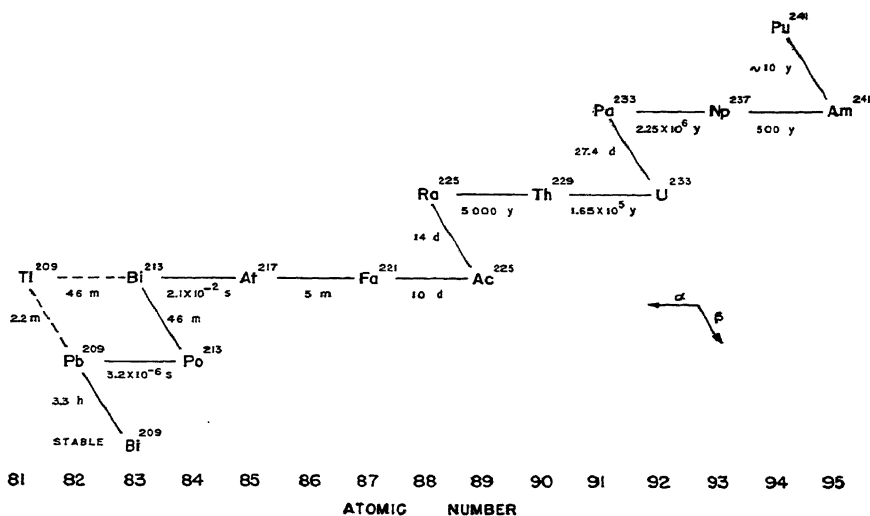


FIG. 53. Neptunium series.

the number of independent series that can be expected to exist. It is seen from Figs. 50, 51, and 52, that the mass number of each member of the uranium-radium series is represented by  $4n + 2$ , where  $n$  is an integer. Similarly, the members of the other two natural series are represented by  $4n + 3$  and  $4n$ , respectively. The new series, it is seen from Fig. 53, is the

TABLE 27. THE NATURAL RADIOACTIVE SERIES AND ALPHA PARTICLES

Isotope	Symbol	A	Z	Half-life	Range, cm in air	Energy, mev	Velocity $3 \times 10^{10}$ cm/sec	Disinte- gration energy, mev	Relative intensity
Uranium-radium Series									
Uranium I	U1	238	92	$4.55 \times 10^9$ y	2.653	4.23	0.0456	4.15	
Uranium X <sub>1</sub>	UX <sub>1</sub>	234	90	24 d	.....	$\beta^-$			
Uranium X <sub>2</sub>	UX <sub>2</sub>	234	91	1.14 m	.....	$\beta^-$			
Uranium Z	UZ	234	91	6.7 h	.....	$\beta^-$			
Uranium II	UII	234	92	$2.3 \times 10^5$ y	3.211	4.76	0.0479	4.7	
Ionium	Io	230	90	$8.2 \times 10^4$ y	(2.85)	4.66	0.0485		
Radium	Ra	226	88	1,600 y	3.13	4.791	0.0500		
Radon	Rn	222	86	3.823 d	4.051	5.4860	0.0540	5.5886	
Radium A	RaA	218	84	3.05 m	4.657	5.9981	0.0565	6.1123	
Radium B	RaB	214	82	26.8 m	.....	$\beta^-$			
Radium C	RaC	214	83	19.72 m	4.039	5.506	0.0544	5.611	94
	RaC'	214	84	$1.5 \times 10^{-4}$ s	3.969	5.445	0.0539	5.549	113
					6.908	7.6802	0.064	7.829	10 <sup>6</sup>
					7.792	8.2769	0.0665	8.437	0.43
						8.941	0.0691	9.112	(0.45)
					9.04	9.0655	0.0696	9.241	22
						9.315	0.0705	9.493	0.38
					9.724	9.4877	0.0711	9.673	1.35
						9.660	0.0718	9.844	0.35
						9.781	0.0723	9.968	1.06
						9.908	0.0728	10.097	0.36
	RaC''	210	81	1.32 m	10.077	0.0734	10.269	1.67	
					10.149	0.0736	10.342	0.38	
					10.329	0.0742	10.526	1.12	
					11.51	10.5052	0.0749	10.709	0.23
					11.580	10.5379			
					.....	$\beta^-$			
					.....	$\beta^-$			
					.....	$\beta^-$			
					3.845	3.685	.....	5.403	21
					(mean)				
Radium D	RaD	210	82	22.2 y	.....	$\beta^-$			
Radium E	RaE	210	83	4.975 d	.....	$\beta^-$			
Radium F	RaF	210	84	139.5 d	.....	$\beta^-$			
(Polonium)	(Po)				.....	$\beta^-$			
					3.890	.....	.....	.....	18
					4.016	.....	.....	.....	19
					4.111	.....	.....	.....	48
					4.303	.....	.....	.....	43
					4.449	.....	.....	.....	79
					4.640	.....	.....	.....	70
					4.749	.....	.....	.....	64
					4.838	.....	.....	.....	49
					4.901	.....	.....	.....	84
					5.065	.....	.....	.....	(126)
					5.113	.....	.....	.....	(96)
					5.2984	0.0523	.....	.....	10 <sup>6</sup>
					5.303	.....	.....	.....	
Radium G	RaG	206	82	stable					
(Lead)	(Pb)								
Actinium-uranium Series									
Actinium-uranium	AcU	235	92	$7.07 \times 10^8$ y	.....	4.52			
Uranium Y	UY	231	90	24.64 h	.....	$\beta^-$			

TABLE 27. THE NATURAL RADIOACTIVE SERIES AND ALPHA PARTICLES—(Continued)

Isotope	Symbol	Half-life	Range, cm in air	Energy, mev	Velocity $3 \times 10^{10}$ cm/sec	Disinte- gration energy, mev	Relative intensity
Actinium-uranium Series—(Continued)							
Protactinium	Pa	231 91	$3.2 \times 10^4$ y	3.20 3.23 3.511	4.69 4.72 5.00	..... ..... .....	4.77 4.80 5.09
Actinium	Ac	227 89	21.7 y	.....	5.0	.....	.....
→ Radioactinium	RdAc	227 90	18.9 d	.....	6.051 6.019 5.990 5.968 5.924 (4.36) 5.870 5.817 5.766 5.744 5.719 5.674	0.0569 0.0567 0.0566 0.0565 0.0563 0.0560 0.0557 0.0555 0.0554 0.0553 0.0550	6.159 6.127 6.097 6.075 6.030 5.975 5.921 5.869 5.847 5.822 5.776
→ Actinium K	AcK	223 87	21 m	.....	$\beta^-$	.....	80 15 100 15 5 10 5 80 15 60 10
Actinium X ←	AcX	223 88	11.2 d	..... (4.17)	5.719 5.607 5.533	0.0552 0.0547 0.0544	5.823 5.709 5.634
Actinon	An	219 86	3.92 s	5.692 5.240 5.14	6.8235 6.561 6.436	0.0603 0.0592 0.0586	6.953 6.683 6.556
Actinium A	AcA	215 84	$1.83 \times 10^{-3}$ s	6.457	7.365	0.0627	.....
Actinium B	AcB	211 82	36 m	.....	$\beta^-$	.....	.....
Actinium C	AcC	211 83	2.16 m	5.429 4.984	6.618 6.272	0.0594 0.0578	6.739 6.383
→ Actinium C'	AcC'	211 84	$2.0 \times 10^{-3}$ s	6.555	7.680	0.0630	100 19
→ Actinium C''	AcC''	207 81	4.71 m	.....	$\beta^-$	.....	.....
Actinium D (Lead)	AcD (Pb)	207 82	stable	.....	.....	.....	.....
Thorium Series							
Thorium	Th	232 90	$1.389 \times 10^{10}$ y	2.80	4.20	0.0481	4.34
Mesothorium 1	MsTh1	228 88	6.7 y	.....	$\beta^-$	.....	.....
Mesothorium 2	MsTh2	228 89	6.13 h	.....	$\beta^-$	.....	.....
Radiothorium	RdTh	228 90	1.90 y	(3.67)	5.420 5.335	(0.0527) .....	5.517 5.431
Thorium X	ThX	224 88	3.64 d	4.08	5.6825	0.0546	5.7858
Thoron	Th	220 86	54.50 s	5.004	6.2818	0.0579	6.399
Thorium A	ThA	216 84	0.145 s	5.638	6.7744	0.0602	6.903
Thorium B	ThB	212 82	10.6 h	.....	$\beta^-$	.....	.....
Thorium C	ThC	212 83	60.6 m	.....	6.083	0.0570	6.200
							27.2

TABLE 27. THE NATURAL RADIOACTIVE SERIES AND ALPHA PARTICLES—(Continued)

Isotope	Symbol	A	Z	Half-life	Range, cm in air	Energy, mev	Velocity $3 \times 10^{10}$ cm/sec	Disinte- gration energy, mev	Relative intensity
Thorium Series—(Continued)									
						6.044	0.0535	6.160	69.8
					(4.730) mean	5.762	0.0555	5.872	1.8
						5.620	0.0548	5.728	0.16
						5.601	0.0544	5.708	1.10
	→ Thorium C'	ThC'	212 84	$3 \times 10^{-7}$ s	8.570	8.7759	0.0684	8.947	10 <sup>4</sup>
					9.687	9.491	0.0712	9.673	34
→ Thorium C''	ThC''	208 81		3.1 m	11.543	10.541	0.0750	10.744	190
					.....	$\beta^-$			
Thorium D (Lead)	ThD (Pb)	208 82		Stable					
Neptunium Series*									
Plutonium	Pu	241 94		.....	.....	$\beta^-$			
Americium	Am	241 95		500 y	.....	$\alpha$			
Neptunium	Np	237 93		$2.25 \times 10^6$ y	.....	$\alpha$			
Protactinium	Pa	233 91		27.4 d	.....	$\beta^-$			
Uranium	U	233 92		$1.63 \times 10^8$ y	.....	4.825			
Thorium	Th	229 90		$5 \times 10^4$ y	.....	$\sim 5$			
Radium	Ra	225 88		14 d	.....	$\beta^-$			
Actinium	Ac	225 89		10 d	.....	5.801			
Francium	Fa	221 87		5 m	.....	6.31			
Astatine	At	217 85		$2.1 \times 10^{-2}$ s	.....	7.023			
Bismuth	Bi	213 83		46 m	.....	5.86			
→ Polonium	Po	213 84		$3.2 \times 10^{-8}$ s	.....	8.336			
→ (Thallium)	Tl	209 81							
Lead	Pb	209 82		3.3 h	.....	$\beta^-$			
Bismuth	Bi	209 83		Stable					

\* Recent data have made it possible to construct a fourth natural radioactive series which has the form  $(4n + 1)$ . Its probable members and arrangement are given above as reported by Hageman, F., L. I. Katzin, M. H. Studier, A. Ghiorso, and G. T. Seaborg, *Phys. Rev.*, **72**, 253 (1947), and by English, A. C., T. E. Cranshaw, P. Demers, J. A. Harvey, E. P. Hinks, J. V. Jelley, and A. N. May, *Phys. Rev.*, **72**, 253 (1947). Thallium 209 is the only member that has not been positively identified. The name "Neptunium Series," after the longest lived member, was proposed in accordance with the practice for naming the other three series.

hitherto missing  $4n + 1$  series. Additional radioisotopes that may be found should belong to existing series. For example, mass number  $4n + 5$  would be merely an existing member or a branch of the  $4n + 1$  series.

Figures 50 to 53 show the order and the half-lives of members of the four series. In the scheme used here, alpha decay corresponds to a displacement of two units to the left, whereas beta decay appears as a displacement of

one unit to the right and diagonally downward. The ordinate of each member, actually the mass number, is represented by the vertical scale  $2Z + Y$  where  $Z$  is the atomic number and  $Y$  is an integer. More detailed data concerning the energies and ranges of alpha particles emitted from the members of the series are given in Table 27.

#### 7.12. Seaborg and Perlman Table of Isotopes.<sup>1</sup>

The following table represents a complete list of all the artificial and natural radioactive isotopes and stable isotopes, together with a number of their important features covering information available by approximately October, 1948, through publications, private communications, and almost all of the restricted distribution reports of the U. S. Atomic Energy Commission, the former "Manhattan District," U. S. Army Corps of Engineers, and the corresponding offices of Great Britain and Canada. With very few exceptions, the criterion for listing a radioactive isotope has been the actual observation of its radiation. A somewhat more extensive treatment of fission product data available up to August, 1946, may be found in a Plutonium Project compilation, "Nuclei formed in fission," *J. Am. Chem. Soc.*, **68**, 2411 (1946).

The first column lists the atomic numbers and mass numbers of the isotopes. The superscript "*m*" following the mass number denotes a metastable isomer of measured half-life of either a stable or unstable ground state, but the isomeric transition need not have been observed.

In the second column headed "Class" the degree of certainty of each isotopic assignment is indicated with a letter according to the following code:

*A* = isotope certain (mass number and element certain)

*B* = isotope probable, element certain

*C* = one of few isotopes, element certain

*D* = element certain

*E* = element probable

*F* = insufficient evidence

In most cases the class is determined by evaluating the uniqueness of the assignment through chemical separation, reaction type and yield considerations, genetic relationships, and type of radiation. In a few cases newer techniques have been used. The term "m.s." in the second column refers to the identification of the mass number by means of a mass spectrograph,

<sup>1</sup> The table of isotopes presented on pp. 187-207 is reprinted from *Rev. Mod. Phys.*, **20**, 585 (1949) by permission of Professors G. T. Seaborg and I. Perlman and the publisher. It was found necessary in the interest of keeping the present volume within reasonable size to omit the exhaustive list of references contained in the original table. Aside from this omission, however, the table is complete in all other details. The explanatory text on this and the succeeding pages is also printed verbatim, except for deleted references and acknowledgments, from the same publication.

and "res.n.act." (resonance neutron activation) refers to the identification of a nuclear isomer by observing both isomers upon irradiation with filtered neutrons. With the mass spectrographic assignment of mass numbers there are some instances in which the mass number is known with greater certainty than the element. Such cases are assigned the appropriate code letter such as "*E*" followed by "m.s."

The per cent abundance of the stable isotopes is listed in column three.

The fourth column lists the type of radiation, with the following meaning for the symbols:

$\beta^-$  = negative beta particles (negatrons)

$\beta^+$  = positive beta particles (positrons)

$\gamma$  = gamma rays

$\alpha$  = alpha particles

$n$  = neutrons

$e^-$  = internal-conversion electrons

K = K-electron capture (or in more general terms, orbital electron capture)

I.T. = isomeric transition (transition from upper to lower isomeric state)

In the cases where it is certain that no gamma rays are emitted, this fact is expressed explicitly in column seven by the term "No  $\gamma$ ." Annihilation gamma rays and x-rays are not listed. It may be assumed that x-rays have been observed or actually identified in almost all cases of orbital electron capture listed.

The half-life is given in the fifth column. In most cases the determination is direct, either by measuring the decay rate, by weighing a long-lived isotope of known purity, or by comparing the activity with that of a genetically related isotope of known half-life. A number of half-lives are known only from the yield of activity resulting from a nuclear reaction of known or estimated cross section. Half-lives estimated in this manner are indicated by the term "yield." Usually for the cases where more than one value for the half-life has been reported, an attempt has been made to list the best value (an experimental value thought to be taken under the most favorable conditions) rather than a mean value; more than one value is listed where a choice does not seem obvious. Among the natural radioactivities an average value is often used which was taken from an international committee summary report.

In the columns headed "Energy of radiation," the energy value is followed by a description of the method used for the energy determination. The beta-particle energies correspond to the observed upper limits of the spectra; in those cases where only the Konopinski-Uhlenbeck extrapolated value has been reported, this is listed, followed by the designation "K.U." For alpha particles reported only by a range the "mean range in air" vs. energy relation-

ship of Holloway and Livingston was used. The methods used for the determination of the energy of the particles (alpha and beta) are described in each case with the aid of the following symbols:

abs. = absorption

cl. ch. = cloud chamber (with magnetic field in case of beta particles)

spect. = magnetic deflection (magnetic spectrograph or spectrometer or counter with magnetic field)

calor. = calorimetric measurements

ion. ch. = measurement of pulse sizes in ionization chamber or proportional counter

coincid. abs. = beta- and gamma-coincidence counters with absorbers

coincid. = beta- and gamma-coincidence counters (for information on decay scheme—data not necessarily used in the table)

spect. coincid. = coincidence counters arranged with a magnetic field

The alpha-particle energies listed, where more than a single group exists in high abundance, include the group of highest energy and those groups with abundance greater than 10 per cent. Conversion electron energies are listed only when it is not known in which shell internal conversion takes place or when no attempt was made to relate the electrons with observed or unobservable gamma rays; in all other cases entries are made in the column for gamma rays.

The symbols used to describe the methods employed for the determination of gamma-ray energies have the following meaning:

abs. = absorption

cl. ch. recoil = secondary electrons in cloud chamber with magnetic field

cl. ch. pair = positron-electron pairs in cloud chamber with magnetic field

coincid. abs. = secondary electrons with coincidence counters and absorbers

spect. conv. = internal-conversion electrons with magnetic spectrograph or spectrometer

spect. = secondary electrons with magnetic spectrograph or spectrometer

cryst. spect. = direct measurement of gamma-ray energy by diffraction in a crystal

abs. of  $e^-$  = absorption of internal-conversion electrons

abs. sec.  $e^-$  = absorption of secondary electrons;

coincid. = measurements with gamma-gamma-coincidence counters (for information on decay scheme—data not necessarily used in the table)

Be- $\gamma$ - $n$  reaction = measurement of neutron energy from Be- $\gamma$ - $n$  reaction

D- $\gamma$ - $n$  reaction = measurement of neutron energy from D- $\gamma$ - $n$  reaction

When internal-conversion electrons are emitted, the energy listed in this column is always that of the corresponding gamma-ray transition. Only

the main gamma rays are listed for the natural radioactive isotopes. In a few instances in which a very short-lived metastable state has been identified as the daughter of the isotope in question, the gamma rays of the daughter may be listed for both parent and daughter.

When a semicolon is used, it means that the values listed on each side of it are independent determinations of the same item, e.g., independent determinations of the half-life or of the energy of the radiation of a radioactivity. In another usage the semicolon separates the symbols in the "type of radiation" columns when there is more than one type of decay ( $\beta^-$ ,  $\beta^+$ ,  $\alpha$ , K, or I.T.) for the radioactivity.

The observed nuclear reactions (giving the target element, projectile, and outgoing particle, in order) by which the radioactive isotopes are formed are listed in the last column ( $p$  = proton,  $n$  = neutron,  $\alpha$  = alpha particle,  $d$  = deuteron,  $t$  = tritium or triton ( $H^3$ ),  $\gamma$  = gamma ray,  $e^-$  = electron). In cases in which the target material is not the naturally occurring element, but one enriched or depleted in a particular isotope, that isotope is indicated. No means for identifying the source or energy of the projectile is given. For example, deuterons varying from low energies to 200 Mev have been used. In many cases, with high-energy projectiles, multiple particles are ejected. A reaction such as  $(d-\alpha p 2n)$  is a formal presentation showing what the outgoing particles might be and does not mean that the order of leaving the nucleus was determined nor that the  $\alpha$ ,  $p$ , and  $n$  were identified.

In some cases where the path for reaching the product nucleus can even less definitely be stated the reaction is presented in the form  $(d-3z10a)$  where "3z" indicates that the product nucleus is lower in atomic number than the compound nucleus by three units and "10a" means that it is lower in mass number by 10 units. Where the same isotope has been made by spallation of various target elements with high-energy particles, this is indicated by the symbol "spal." followed by the symbols for the target elements.

Stable product nuclei which have been identified by means of the mass spectrograph are indicated by "m.s." The neutron-induced fission reactions of the heavy elements are designated by such symbols as U- $n$ , Th- $n$ , Pu- $n$ , and Pa- $n$ , while the gamma ray, deuteron, and alpha-particle-induced fission reactions are designated by symbols such as U- $\gamma$ , U- $d$ , and U- $\alpha$ . Usually, but not always, "U- $n$ " will mean the slow neutron fission of  $U^{235}$  while "U- $d$ " or "U- $\alpha$ " designated fission products arise from  $U^{238}$ . In this last column the method of production for each radioactive fission product is described by these symbols (U- $n$ , etc.) together with the designation of its radioactive parent and its radioactive daughter when these are known. Similarly, for the radioactivities of the heavy natural and artificial families there are listed the immediate parent and daughter isotopes. The natural radioactivities without parents are listed as produced by a "natural source."



Isotope Z      A	Class	Per cent abundance	Type of radiation	Half-life	Energy of radiation, Mev		Produced by
					Particles	$\gamma$ -rays	
1 H <sup>1</sup> H <sup>2</sup> H <sup>3</sup>	A	99.9844 0.0156	$\beta^-$	12.1 yr; 10.7 yr	0.0185 ion.ch.; 0.0169 ion.ch.; 0.015 abs., cl.ch.; 0.011 abs., cl.ch.	No $\gamma$	D-n- $\gamma$ D-d- $p$ He <sup>3</sup> -n- $p$ Li-n-t Be-d-t B-n-t N-n-t
2 He <sup>3</sup> He <sup>4</sup> He <sup>6</sup>	A	$1.3 \times 10^{-4}$ 99.9999	$\beta^-$	0.89 sec; 0.8 sec; 0.85 sec	3.7 cl.ch.; 3.7 abs. Al; 3.5 abs. Al	No $\gamma$	Li- $\gamma$ - $p$ Li-n- $p$ Be-n- $\alpha$
3 Li <sup>6</sup> Li <sup>7</sup> Li <sup>8</sup>	A	7.39 92.61	$\beta^-$ , $2\alpha$	0.89 sec; 0.88 sec	12( $\beta^-$ ) cl. ch.; 12( $\beta^-$ ) abs. Al; distribu- tion, mean at 2.0( $\alpha$ )	No $\gamma$	Li-d- $p$ Li-n- $\gamma$ Li <sup>7</sup> -n- $\gamma$ Be- $\gamma$ - $p$ B-n- $\alpha$
4 Be <sup>7</sup>	A		K, $\gamma$	52.9 days; 43 days		0.485 spect. 0.485 coincid. abs.; 0.476 abs. Pb; 0.453 spect.; 0.474 spect.	Li-d-n Li-p-n B-p- $\alpha$ B-d- $\alpha$ n
Be <sup>8</sup>	A		$2\alpha$	$10^{-14}$ — $10^{-17}$ sec calc.	0.055 (each $\alpha$ in cen- ter of mass system) ion.ch.		Be- $\gamma$ -n
Be <sup>9</sup> Be <sup>10</sup>	A m.s.	100	$\beta^-$	$2.5 \times 10^4$ yr; $2.9 \times 10^6$ yr yield	0.560 abs. Al; 0.58 abs. Al; 0.65 abs. Al	No $\gamma$	Be-d- $p$ Be-n- $\gamma$ B-n- $p$ C-n- $\alpha$
5 B <sup>10</sup> B <sup>11</sup> B <sup>12</sup>	A	18.83 81.17	$\beta^-$	0.027 sec; 0.022 sec	12 cl.ch.		B-d- $p$ N <sup>14</sup> -n- $\alpha$
6 C <sup>10</sup>	B		$\beta^+$	20 sec	$\sim 2$ abs.		B-p-n B <sup>10</sup> -p-n

Isotope Z      A	Class	Per cent abundance	Type of radiation	Half-life	Energy of radiation, Mev		Produced by
					Particles	$\gamma$ -rays	
6 C <sup>11</sup>	A		$\beta^+$	20.5 min; 20.0 min	0.95 cl.ch.; 0.99 spect.	No $\gamma$ coincid.	Be- $\alpha$ -2n B-d-n B-p- $\gamma$ B-p-n C- $\gamma$ -n C-n-2n C-d-dn C-p-pn C- $\alpha$ - $\alpha$ n N-p- $\alpha$ N-n-p3n N- $\gamma$ -p2n O- $\gamma$ - $\alpha$ n(?) O-n- $\alpha$ 2n
C <sup>12</sup> C <sup>13</sup> C <sup>14</sup>	A	98.9 1.1	$\beta^-$	5100 yr; 6400 yr; 4700 yr	0.156 spect.; 0.154 abs. Al; 0.154 spect.	No $\gamma$	C-d-p C-n- $\gamma$ N-n-p O-n- $\alpha$
7 N <sup>12</sup>	A		$\beta^+$	9.93 min; 10.13 min	1.24 spect.; 1.25 spect.; 0.92, 1.20 spect.	No $\gamma$ coin- cid.; No $\gamma$ spect.	B- $\alpha$ -n C-d-n C-p- $\gamma$ N-n-2n N-d-t N- $\gamma$ -n O-n-p3n
N <sup>14</sup> N <sup>15</sup> N <sup>16</sup>	A	99.62 0.38	$\beta^-$ , $\gamma$	7.35 sec; 7.5 sec; 7.3 sec; 8 sec	3.5, 10 abs. Al, Cu; 10 cl.ch; 4, 10.3 cl.ch., abs.	6.2, 6.7 abs. sec e-, cl. ch. pair; 4 abs. Pb, Cu; ~ 6 cl.ch. recoil	N-n- $\gamma$ N-d-p O-n-p F-n- $\alpha$
N <sup>17</sup>	A		$\beta^-$ , n	4.14 sec	3.7 ( $\beta^-$ ) O <sup>16</sup> recoil— $\beta^-$ coincid. abs.; 0.9 (mean)(n) O <sup>16</sup> recoil in ion.ch.; 1.0(mean) (n) p re- coil in cl.ch.		Spal.(O,F, N,Mg,Al, Si,P,S,Cl, K)
8 O <sup>14</sup> O <sup>15</sup>	B A		$\beta^+$ $\gamma$ $\beta^+$	76.5 sec 126 sec	1.8 abs. 1.7 cl.ch.	2.3 abs.	N-p-n C- $\alpha$ -n N-d-n N-p- $\gamma$

Isotope Z      A	Class	Per cent abundance	Type of radiation	Half-life	Energy of radiation, Mev		Produced by
					Particles	$\gamma$ -rays	
8 O <sup>15</sup>							O- $\gamma$ - $n$ O- $n$ -2 $n$
O <sup>16</sup>		99.757					
O <sup>17</sup>		0.039					
O <sup>18</sup>		0.204					
O <sup>19</sup>	A		$\beta^-$ , $\gamma$	29.4 sec; 29.5 sec; 27.0 sec	4.5 (30 %), 2.9 (70 %) abs. Al; 4.1 abs.; 3.2 abs. Al	1.6 abs.	O- $n$ - $\gamma$ F- $n$ - $p$
9 F <sup>17</sup>	A		$\beta^+$	70 sec	2.1 cl.ch.		N- $\alpha$ - $n$ O- $d$ - $n$ O- $p$ - $\gamma$ F- $\gamma$ -2 $n$
F <sup>18</sup>	A		$\beta^+$	112 min	0.7 cl.ch.; 0.7 abs. Al; 0.95 (20 %), 0.6 (80 %) cl.ch.	No $\gamma$ ; 1.4 cl.ch. recoil	O- $\alpha$ - $p$ $n$ O- $p$ - $n$ O- $d$ - $n$ O- $t$ - $n$ F- $n$ -2 $n$ F- $d$ - $t$ F- $\gamma$ - $n$ Ne- $d$ - $\alpha$ Na- $\gamma$ - $\alpha$ (?)
F <sup>19</sup>		100					
F <sup>20</sup>	A		$\beta^-$ , $\gamma$	12 sec	5.0 cl.ch.	2.2 cl.ch. recoil	F- $d$ - $p$ F- $n$ - $\gamma$ Na- $n$ - $\alpha$
10 Ne <sup>19</sup>	A		$\beta^+$	20.3 sec	2.20 cl.ch.		F- $p$ - $n$
Ne <sup>20</sup>		90.51					
Ne <sup>21</sup>		0.28					
Ne <sup>22</sup>		9.21					
Ne <sup>23</sup>	A		$\beta^-$	40 sec; 40.7 sec	4.1 abs.		Ne- $d$ - $p$ Na- $n$ - $p$ Mg- $n$ - $\alpha$
11 Na <sup>21</sup>	B			23 sec			Ne- $p$ - $n$ Ne- $d$ - $n$ Mg <sup>24</sup> - $p$ - $\alpha$
Na <sup>22</sup>	A		$\beta^+$ (~100 %), $\gamma$ , no K	2.6 yr; 3.0 yr	0.58 cl.ch.; 0.575 spect.; coincid.	1.3 spect.; 1.30 spect.	F- $\alpha$ - $n$ Ne- $d$ - $n$ Na- $n$ -2 $n$ Mg- $d$ - $\alpha$
Na <sup>23</sup>		100					
Na <sup>24</sup>	A		$\beta^-$ , $\gamma$	14.8 hr	1.390 spect., coincid.; 1.4 spect.	1.380, 2.758 spect.; 1.4, 2.8 spect.; 2.87, 2.74 Be- $\gamma$ - $n$ reaction, D- $\gamma$ - $n$ reaction; 2.56, 2.68,	Na- $d$ - $p$ Na- $n$ - $\gamma$ Mg- $d$ - $\alpha$ Mg- $n$ - $p$ Mg- $\gamma$ - $p$ Al- $n$ - $\alpha$ Al- $d$ - $p$ $\alpha$ Al- $\gamma$ - $n$ 2 $p$ Si- $\gamma$ - $n$ 3 $p$ (?)

Isotope Z      A	Class	Per cent abundance	Type of radiation	Half-life	Energy of radiation, Mev		Produced by
					Particles	$\gamma$ -rays	
11 Na <sup>24</sup>						2.76, 2.89 cl.ch. pair; coincid. abs.	
Na <sup>26</sup>	B		$\beta^-$ , $\gamma$	58.2 sec; 60 sec; 62 sec	3.4 abs. Al; 2.8 abs. Al		Mg- $\gamma$ - $p$ Mg- $n$ - $p$ Al- $\gamma$ -2 $p$
12 Mg <sup>23</sup>	A		$\beta^+$	11.6 sec	2.82 cl.ch.		Na- $p$ - $n$ Mg- $\gamma$ - $n$
Mg <sup>24</sup>		78.60					
Mg <sup>25</sup>		10.11					
Mg <sup>26</sup>		11.29					
Mg <sup>27</sup>	A		$\beta^-$ , $\gamma$	10.2 min; 9.6 min	0.79 (20 %), 1.80 (80 %) spect.; 1.8 cl.ch.; coincid.	1.01, 0.84 spect., coincid.; 0.64, 0.84, 1.02 spect.; 1.05 (single $\gamma$ ) cl.ch. recoil	Mg- $d$ - $p$ Mg- $n$ - $\gamma$ Al- $n$ - $p$
13 Al <sup>25</sup>	A			8 sec; 7.3 sec			Mg <sup>24</sup> - $p$ - $n$
Al <sup>26</sup>	A		$\beta^+$	6 sec; 6.3 sec 7.0 sec	2.99 cl.ch.; 1.8 abs.		Na- $\alpha$ - $n$ Mg- $p$ - $n$ Mg <sup>24</sup> - $p$ - $n$ Mg- $p$ - $\gamma$ Al- $\gamma$ - $n$
Al <sup>27</sup>		100					
Al <sup>28</sup>	A		$\beta^-$ , $\gamma$	2.30 min; 2.4 min	3.01 spect.; 2.75 coincid. abs.; 3.3 cl.ch.; 3.0 cl.ch.; 3.10 abs. Al, coincid.	1.80 abs. sec. $e^-$ ; 1.80 spect.; 1.8 spect.; 2.1 cl.ch. recoil	Mg- $\alpha$ - $p$ Al- $d$ - $p$ Al- $n$ - $\gamma$ Si- $n$ - $p$ Si- $\gamma$ - $p$ P- $n$ - $\alpha$
Al <sup>29</sup>	A		$\beta^-$	6.7 min	2.5 cl.ch. and abs.		Mg- $\alpha$ - $p$ Si- $n$ - $p$ Si- $\gamma$ - $p$ P- $\gamma$ -2 $p$
14 Si <sup>27</sup>	A		$\beta^+$	4.9 sec	3.74 cl.ch.; 3.54 cl.ch.		Al- $p$ - $n$ Mg- $\alpha$ - $n$ Si- $\gamma$ - $n$
Si <sup>28</sup>		92.28					
Si <sup>29</sup>		4.67					
Si <sup>30</sup>		3.05					
Si <sup>31</sup>	A		$\beta^-$	170 min	1.8 cl.ch.	No $\gamma$	Si- $d$ - $p$ Si- $n$ - $\gamma$ P- $n$ - $p$ S- $n$ - $\alpha$

Isotope Z     A	Class	Per cent abundance	Type of radiation	Half-life	Energy of radiation, Mev		Produced by
					Particles	$\gamma$ -rays	
15 P <sup>29</sup>	A	100	$\beta^+$	4.6 sec	3.63 cl.ch.	No $\gamma$	Si-p-n Si-d-n P- $\gamma$ -2n(?)
P <sup>30</sup>	A		$\beta^+$	2.55 min	3.0 cl.ch.; 3.5 spect.		Al- $\alpha$ -n Si-p-n Si-He <sup>2</sup> -p P-n-2n P- $\gamma$ -n S-d- $\alpha$
P <sup>31</sup> P <sup>32</sup>	A		$\beta^-$	14.30 days; 14.07 days	1.712 spect.; 1.69 spect.		Si- $\alpha$ -p P-d-p P-n- $\gamma$ S-n-p S-d- $\alpha$ Cl-n- $\alpha$ Cl-d-p $\alpha$ Cu-d-15z33a
P <sup>34</sup>	B		$\beta^-, \gamma$	12.4 sec	5.1 (75 %), 3.2 (25 %) coincid. abs.; 4.9 abs. Al		S-n-p Cl-n- $\alpha$
16 S <sup>31</sup>	A		$\beta^+$	2.6 sec; 3.2 sec	3.85 cl.ch.		Si- $\alpha$ -n P-p-n S- $\gamma$ -n
S <sup>32</sup> S <sup>33</sup> S <sup>34</sup> S <sup>35</sup>	A	95.06 0.74 4.18	$\beta^-$	87.1 days	0.169 spect.; 0.167 abs. Al; 0.166 spect.; 0.17 abs. Al	2.6 abs.; 2.75 abs. sec. $e^-$	S-n- $\gamma$ S-d-p Cl-n-p Cl-d- $\alpha$
S <sup>36</sup> S <sup>37</sup>	B	0.016	$\beta^-, \gamma$	5.04 min; 5.0 min	4.3 (10 %), 1.6 (90 %); 4, 1.4 abs. Al		S-n- $\gamma$ Cl-n-p
17 Cl <sup>32</sup>	A	75.4	$\beta^+$	2.4 sec; 2.8 sec.	4.13 cl.ch.		S-d-n S-p-n
Cl <sup>34</sup>	A		$\beta^+, \gamma$	33 min	2.5 abs.; 5.1, 2.4 cl.ch.	3.4 cl.ch. recoil	P- $\alpha$ -n S-d-n  S- $\alpha$ -p, n or S- $\alpha$ -d S-l-n Cl-n-2n Cl- $\gamma$ -n
Cl <sup>35</sup>							

Isotope $Z$ $A$	Class	Per cent abundance	Type of radiation	Half-life	Energy of radiation, Mev		Produced by
					Particles	$\gamma$ -rays	
17 $\text{Cl}^{36}$	A		$\beta^+$ ; K; $\beta^-$	$2 \times 10^6$ yr yield $\beta^-$ , $\beta^+$ ; $\sim 10^6$ yr yield; $> 10^3$ yr yield	0.64 ( $\beta^-$ ) abs.; 0.66 ( $\beta^-$ ) abs. Al		Cl-n- $\gamma$ Cl-d- $p$
$\text{Cl}^{37}$ $\text{Cl}^{38}$	A	24.6	$\beta^-$ , $\gamma$	38.5 min; 37 min	1.19 (36 %), 2.70 (11 %), 5.2 (53 %) spect.; 1.1, 2.8, 5.0 spect.; coincid. abs.	1.60 (43 %), 2.12 (57 %) spect.; 1.65, 2.15 spect.	Cl-d- $p$ Cl-n- $\gamma$ K-n- $\alpha$ Cu-d-13s27u
$\text{Cl}^{39}$	B		$\beta^-$	1 hr			Cu-d-13s26a Cu- $\alpha$ -14s28a As-d-17s38a
18 $\text{A}^{35}$	A		$\beta^+$	1.88 sec; 1.84 sec	4.4 cl.ch.		S- $\alpha$ -n Cl-p-n
$\text{A}^{36}$ $\text{A}^{37}$	A	0.307	K	34.1 days		No $\gamma$	S- $\alpha$ -n Cl-d-2n Cl-p-n K-d- $\alpha$ Ca-n- $\alpha$
$\text{A}^{38}$ $\text{A}^{39}$ $\text{A}^{40}$ $\text{A}^{41}$	F A	0.060 99.633	$\beta^-$ $\beta^-$ , $\gamma$	4 min 110 min; 109.4 min	1.18, 2.55 (0.7 %) abs. Al, coincid.; 1.5 cl.ch. (K.U.)	1.37 cl.ch. recoil; 1.3 abs. of $e^-$	K-n-p A-d-p A-n- $\gamma$ K-n-p
19 $\text{K}^{37}$ $\text{K}^{38}$	F A		$\beta^+$ $\beta^+$ , $\gamma$	1.3 sec 7.7 min; 7.5 min	2.53 abs. Al; 2.3 abs.	2.15 co- incid. abs.	K- $\gamma$ -2n Cl- $\alpha$ -n K-n-2n K- $\gamma$ -n Ca-d- $\alpha$
$\text{K}^{39}$ $\text{K}^{40}$	A	93.3 0.011	$\beta^-$ ; K; K/ $\beta^-$ ratio $\sim 0.1, 1.9$ , $> 1$ ; $\gamma$ (14 % of $\beta^-$ )	$1.8 \times 10^9$ yr (uncorr. for K); $1.4 \times 10^9$ yr (uncorr. for K); $1.5 \times 10^9$ yr (uncorr. for K)	1.9 abs. Al; 1.7 cl.ch.; 1.41 abs. Al; 1.35 spect. coincid.	1.54 (with K) coincid.; 1.55 abs. Pb; 1.5 (with K) abs. Cu, Pb, co- incid.	Natural source
$\text{K}^{41}$		6.7					

Isotope Z     A	Class	Per cent abundance	Type of radiation	Half-life	Energy of radiation, Mev		Produced by
					Particles	$\gamma$ -rays	
19 K <sup>42</sup>	A		$\beta^-$ , $\gamma$	12.4 hr; 12.44 hr	2.04(25 %), 3.58(75 %) spect.; ~ 1.8, 3.50 abs. Al, co- incid.; 3.5 cl.ch.	1.4, 2.1 abs. sec. $e^-$ ; 1.51 spect., coincid.	A- $\alpha$ -pn K-d-p K-n- $\gamma$ Ca-n-p Sc-n- $\alpha$
K <sup>43</sup>	B		$\beta^-$ , $\gamma$	22.4 hr	0.24, 0.81 spect.	0.4 abs. Pb	A- $\alpha$ -p
K <sup>43</sup>	D		$\beta^-$	27 min			Ca-n-p
K <sup>43,44</sup>	C		$\beta^-$	18 min			Ca-n-p
20 Ca <sup>39</sup>	F		$\beta^+$	4.5 min			Ca-n-2n(?)
Ca <sup>39</sup>	E			1.06 sec			Ca- $\gamma$ -n
Ca <sup>40</sup>		96.96					
Ca <sup>42</sup>		0.64					
Ca <sup>43</sup>		0.15					
Ca <sup>44</sup>		2.06					
Ca <sup>45</sup>	A		$\beta^-$	152 days; 180 days	0.260 abs. Al; 0.25 spect.; 0.21	No $\gamma$	Ca-n- $\gamma$ Ca-d-p Sc-n-p Ti-n- $\alpha$ Bi-d Sc-d-2p
Ca <sup>46</sup>		0.0033					
Ca <sup>47</sup>	F		$\beta^-$ , $\gamma$	5.8 days	1.1	1.3	Ca-d-p
Ca <sup>48</sup>		0.19					
Ca <sup>48</sup>	A		$\beta^-$ , $\gamma$	2.5 hr	2.3 abs.	0.8 abs. Pb	Ca-d-p Ca-n- $\gamma$
Ca <sup>49</sup>	B		$\beta^-$	30 min			Ca-d-p Ca-n- $\gamma$
21 Sc <sup>41</sup>	A		$\beta^+$	0.87 sec	4.94 cl.ch.		Ca-d-n
Sc <sup>43</sup>	A		$\beta^+$ , $\gamma$	3.92 hr; 4 hr	1.12 abs. Al, spect.; 0.4, 1.4 abs.	1.65 abs. Pb, Cu; 1.0 abs. Pb	Ca- $\alpha$ -p Ca-d-n Ca-p-n Sc-n-2n
Sc <sup>44m</sup>	A		I.T., $e^-$ , $\gamma$	2.44 days; 2.2 days		0.27 spect. conv.; 0.28 abs. of $e^-$	K- $\alpha$ -n Ca-d-n Ca-p-n Sc-n-2n Ti-d- $\alpha$
Sc <sup>44</sup>	A		$\beta^+$ , $\gamma$ , K	3.92 hr; 4.1 hr	1.5 abs., spect.; 1.33 abs. Al	1.33 abs. Pb, Cu, Al	K- $\alpha$ -n Ca-d-n Ca-p-n Sc-n-2n Sc- $\gamma$ -n Sc <sup>44m</sup> I.T. Ti-d- $\alpha$
Sc <sup>45</sup>		100					
Sc <sup>46m</sup>	A		I.T., $\gamma$ , $e^-$	20 sec		0.18 abs., abs. of $e^-$	Sc-n- $\gamma$
	res.n.act.						

Isotope $Z \quad A$	Class	Per cent abundance	Type of radiation	Half-life	Energy of radiation, Mev		Produced by
					Particles	$\gamma$ -rays	
21 Sc <sup>46</sup>	A		$\beta^-$ , $\gamma$ , K	85 days	0.36( $\beta^-$ ) spect.; 0.26( $\beta^-$ ) abs. Al; coincid.; 0.4( $\beta^-$ ) abs. Al; 0.26, 1.5( $\beta^-$ ) abs.; 1.49( $\beta^-$ ) (weak) spect.	0.88, 1.12 spect.; coincid.; 1.25 abs. Pb; 1.5 abs. Pb; 1.4 abs. Pb	Ca- $\alpha$ -p Sc-d-p Sc-n- $\gamma$ Ti-d- $\alpha$ Ti-n-p
Sc <sup>47</sup>	B		$\beta^-$	3.4 days	0.46 abs. Al	No $\gamma$ (?)	Ca- $\alpha$ -p Ca-d-n Ca-p- $\gamma$ Ti-n-p
Sc <sup>48</sup>	A		$\beta^-$ , $\gamma$ , K(?)	44 hr	0.64 spect.; 0.57 abs. Al	0.98, 1.33 spect.; 1.35 spect.; 1.33 abs. Pb	Ca-p-n Ca-d-2n Ti-n-p Ti-d- $\alpha$ V-n- $\alpha$
Sc <sup>49</sup>	A		$\beta^-$	57 min	1.8 abs.	No $\gamma$	Ca-d-n Ti-n-p Ti- $\gamma$ -p Ca <sup>49</sup> (2.5 hr) $\beta^-$ -decay Ca <sup>49</sup> (30 min) $\beta^-$ -decay
22 Ti <sup>42</sup>	E			0.58 sec			Ca- $\alpha$ -n
Ti <sup>46</sup>	A		$\beta^+$	3.08 hr	1.2 cl.ch.		Ca- $\alpha$ -n Sc-p-n Sc-d-2n Ti-n-2n Ti- $\gamma$ -n Cu-d-8s20a Sc-p-n
Ti <sup>45</sup>	D			21 days			
Ti <sup>46</sup>		7.95					
Ti <sup>47</sup>		7.75					
Ti <sup>48</sup>		73.45					
Ti <sup>49</sup>		5.51					
Ti <sup>50</sup>		5.34					
Ti <sup>51m</sup>	A		$\beta^-$ , $\gamma$	6 min	1.6 abs.		Ti-d-p Ti-n- $\gamma$
Ti <sup>61</sup>	A		$\beta^-$ , $\gamma$	72 days	0.45 abs. Al; 0.36 abs.	1.0 coincid. abs.; 1.02 coincid. abs.	Ti-d-p Ti-n- $\gamma$ Cu-d-8s14a
23 V <sup>47</sup>	B		$\beta^+$	33 min	1.9 abs.		Ti-d-n Ti-p-n



Isotope Z      A	Class	Per cent abundance	Type of radiation	Half-life	Energy of radiation, Mev		Produced by
					Particles	$\gamma$ -rays	
23 V <sup>48</sup>	A	100	$\beta^-$ , K, $\gamma$ ; $\beta^+$ (58 $\epsilon_i$ ), K (42 $\epsilon_i$ )	16 days	0.72 spect.; 1.0 cl.ch.; 0.58	0.98, 1.33 spect.; 1.05 cl.ch. recoil; 1.50 abs. Pb	Sc- $\alpha$ -n Ti-d-n Ti-p-n Cr-d- $\alpha$ Cu-d-7 $\pm$ 17 $\alpha$ Ti-d-n
V <sup>49</sup>	B		K	600 days	No $\beta$ or $e^-$	No $\gamma$	
V <sup>51</sup>	A		$\beta^-$ , $\gamma$	3.74 min; 3.9 min	2.05 abs.; 2.65 cl.ch.	1.46 abs. Pb, Fe, Cu; 1.3 abs. Pb	V-n- $\gamma$ V-d-p Cr-n-p Cr- $\gamma$ -p Mn-n- $\alpha$
V <sup>52</sup>							
24 Cr <sup>49</sup>	A	4.49	$\beta^+$ , $\gamma$	41.9 min; 45 min	1.45 abs., cl.ch.	0.18, 1.55 abs. Pb	Ti- $\alpha$ -n Cr-n-2n Cr- $\gamma$ -n Cu-d-6 $\pm$ 16 $\alpha$ or Cu-d-6 $\pm$ 18 $\alpha$
Cr <sup>50</sup>	A		K, $\gamma$ , $e^-$ ; no $\beta^+$	26.5 days		0.32 (single) spect. conv.; 0.320 (sin- gle) spect.; 0.330, 0.237 abs. of $e^-$	Ti- $\alpha$ -n V-p-n Cr-d-p Cr-n- $\gamma$ Cr-n-2n Cu-d-6 $\pm$ 14 $\alpha$ As-d-10 $\pm$ 26 $\alpha$
Cr <sup>51</sup>							
Cr <sup>52</sup>							
Cr <sup>54</sup>	B	9.43		1.3 hr; 1.6-2.3 hr			Cr-n- $\gamma$ Cr-d-p
Cr <sup>56</sup>		2.30					
25 Mn <sup>51</sup>	A		$\beta^+$	46 min	2.0 abs.		Cr-d-n Cr-p- $\gamma$ Cu-d-5 $\pm$ 14 $\alpha$
Mn <sup>52m</sup>	A		$\beta^+$ , $\gamma$ ; I.T.(?) (0.05 %)	21 min	2.66 spect.; 2.2 cl.ch.	1.46 spect., coincid.; 1.2; 0.39 (I.T.?) spect. conv.	Fe-d- $\alpha$ Cr-p-n Fe <sup>52</sup> $\beta^+$ -de- cay
Mn <sup>53</sup>	A		$\beta^+$ (35 %), K (65 %)	5.8 days; 6.5 days	0.58 spect.; 0.77 cl.ch.; 0.75 abs. Al	1.0; 0.73, 0.94, 1.46 spect., coincid. abs.	Cr-p-n Cr-d-2n Fe-d- $\alpha$ Cu-d-5 $\pm$ 13 $\alpha$ or Cu-d-5 $\pm$ 15 $\alpha$ As-d-9 $\pm$ 25 $\alpha$
Mn <sup>54</sup>	A		K, $\gamma$	310 days		0.835 spect., coincid.; 0.85 abs. Pb	V- $\alpha$ -n Cr-d-n Cr-p-n Fe-d- $\alpha$

Isotope Z      A	Class	Per cent abundance	Type of radiation	Half-life	Energy of radiation, Mev		Produced by
					Particles	$\gamma$ -rays	
25 Mn <sup>55</sup> Mn <sup>56</sup>	A	100	$\beta^-$ , $\gamma$	2.59 hr	0.75, 1.05, 2.86 spect., coincid.; 1.04, 2.88 spect.; 0.75 (20 %), 1.04 (30 %), 2.81 (50 %) spect.	2.06 (20 %), 1.77 (30 %), 0.822 (~ 100 %) spect.; 0.845, 1.81, 2.13 spect.; 2.7 ( $\leq$ 1 %) D- $\gamma$ -n reaction	Cr- $\alpha$ -p Mn-n- $\gamma$ Mn-d-p Fe-d- $\alpha$ Fe-n-p Fe- $\gamma$ -p Co-n- $\alpha$ Cu-d-p2 $\alpha$ or Cu-d- p2 $\alpha$ 2n As-d-9s21a
26 Fe <sup>52</sup>	A		$\beta^+$	7.8 hr	0.55 abs. Al		Cu-d-4s13a or Cu-d- 4s15a, parent of Mn <sup>52m</sup>
Fe <sup>52</sup>	A		$\beta^+$	8.9 min			Cr- $\alpha$ -n Fe-n-2n Fe- $\gamma$ -n Cu-d-4s12a or Cu-d-4s14a
Fe <sup>54</sup> Fe <sup>55</sup>	A	5.81	K, no $e^-$ , no $\beta^+$	~ 4 yr		No $\gamma$	Mn-d-2n Mn-p-n Fe-d-p Co <sup>55</sup> $\beta^+$ - decay
Fe <sup>56</sup> Fe <sup>57</sup> Fe <sup>58</sup> Fe <sup>59</sup>	A	91.64 2.21 0.34	$\beta^-$ , $\gamma$	46.3 days; 45.5 days; 47 days	0.26, 0.46 spect., coincid. abs.	1.10, 1.30 spect.	Fe-d-p Fe-n- $\gamma$ Co-n-p Co-d-2p Cu-d- $\alpha$ 2p or Cu-d-2 $\alpha$ As-d-8s18a Bi-d
27 Co <sup>55</sup>	A		$\beta^+$ , $\gamma$	18.2 hr	1.50 spect.	0.16, 0.21, 0.8, 1.2 cl.ch. recoil	Fe-d-n Fe-p- $\gamma$ Cu-d-3s10a or Cu-d-3s12a As-d-7s22a
Co <sup>56</sup>	A		$\beta^+$ , $\gamma$ , K	72 days	1.50 spect., coincid.; 1.2 abs., cl.ch., co- incid.	0.845, 1.26, 1.74, 2.01, 2.55, 3.25 spect., co- incid.; 1.7	Fe-d-2n Fe- $\alpha$ -np Ni-d- $\alpha$ Cu-d-3s9a or

Isotope Z      A	Class	Per cent abundance	Type of radiation	Half-life	Energy of radiation, Mev		Produced by
					Particles	$\gamma$ -rays	
27 Co <sup>56</sup>						abs. Pb, coincid.; 1.05 abs. Pb	Cu-d-3 $\pm$ 11a
Co <sup>57</sup>	A		K, $\gamma$ , $e^-$ ; $\beta^+$	270 days	0.26( $\beta^+$ )	0.117, 0.130, 0.202, 0.215 spect.	Fe-d-n Fe-p- $\gamma$
Co <sup>58</sup>	A		$\beta^+$ , $\gamma$ (15 %); K, $\gamma$ (85 %)	72 days	0.470 spect.; coincid.; 0.4 abs.	0.805 spect., coincid.; 0.6 abs. Pb	Mn- $\alpha$ -n Fe-d-n Fe-p-n Fe- $\alpha$ -n-p Fe-p- $\gamma$ Ni-d- $\alpha$ Ni-n-p Cu-d- $\alpha$ -p2n or Cu-d- $\alpha$ -p4n
Co <sup>59</sup> Co <sup>60</sup>	A	100	$\beta^-$ , $\gamma$	5.3 yr	0.31 spect. coincid. abs.; 0.23 spect.; 0.310 spect.	1.16, 1.32 spect.; 1.16, 1.30 spect.; 1.10, 1.30 spect., coincid.	Co-d-p Co-n- $\gamma$ Co <sup>60m</sup> I.T. Ni-d- $\alpha$ Cu-n- $\alpha$
Co <sup>60m</sup>	A		I.T., $\gamma$ , $e^-$ ( $> 90$ %); $\beta^-$ , $\gamma$ ( $< 10$ %)	10.7 min	1.35( $\beta^-$ ) spect.; 1.25( $\beta^-$ ) spect.; 1.56( $\beta^-$ ) spect.	0.056(I.T.) spect. conv.; 1.5 (with $\beta^-$ ) abs. Pb; 1.32 (with $\beta^-$ ) spect.	Co-n- $\gamma$ Co-d-p Ni-n-p
Co <sup>61</sup>	A m.s.		$\beta^-$	1.75 hr	1.1 abs. Al	No $\gamma$	Co-t-p Ni- $\gamma$ -p Ni-d- $\alpha$ n Ni- $\alpha$ -p- $\alpha$ Ni- $\alpha$ -n-p Cu-n-n $\alpha$ Cu- $\gamma$ -2p Cu-d- $\alpha$ -p As-d-7 $\pm$ 16a
Co <sup>62</sup>	B		$\beta^-$ , $\gamma$	13.8 min	2.5 abs. Al, coincid.	1.3 abs. Pb	Ni- $\alpha$ -n-p Cu-n- $\alpha$ Cu-d- $\alpha$ -p
28 Ni <sup>57</sup>	A		$\beta^+$	36 hr; 34 hr	0.67 abs.		Fe- $\alpha$ -n Ni-n-2n Ni- $\gamma$ -n Cu-d-2 $\pm$ 8a or Cu-d-2 $\pm$ 10a As-d-6 $\pm$ 20a

Isotope $Z$ $A$	Class	Per cent abundance	Type of radiation	Half-life	Energy of radiation, Mev		Produced by
					Particles	$\gamma$ -rays	
28 Ni <sup>58</sup> Ni <sup>59</sup>	B	67.76	K, no $\beta^-$	$5 \times 10^4$ yr yield; 16 yr yield	$\sim 0.05$ abs. Al		Fe- $\alpha$ -n Ni-n- $\gamma$ Ni-d-p Co-d-2n
Ni <sup>60</sup> Ni <sup>61</sup> Ni <sup>62</sup> Ni <sup>63</sup>	B	26.16 1.25 3.66	$\beta^-$	300 yr yield; long yield	0.05 abs. Al; abs. A, Al		Ni-n- $\gamma$ Ni <sup>62</sup> -n- $\gamma$
Ni <sup>64</sup> Ni <sup>65</sup>	A	1.16	$\beta^-$ , $\gamma$	2.6 hr	1.9 abs. Al	1.1 abs. Pb; 0.280, 0.65, 0.93 spect.	Ni-d-p Ni-n- $\gamma$ Ni <sup>64</sup> -n- $\gamma$ Cu-n-p Zn-n- $\alpha$ Cu <sup>65</sup> -n-p Cu-d-2p As-d-6s12a As-d-6s11a Bi-d, parent of Cu <sup>66</sup>
Ni <sup>66</sup>	A		$\beta^-$	56 hr			
29 Cu <sup>68</sup>	D		$\beta^+$	7.9 min; 10 min			Ni-p-n
Cu <sup>68</sup>	B			3 sec			Ni-p-n Ni <sup>68</sup> -p-n
Cu <sup>69</sup> Cu <sup>70</sup>	E A m.s.		$\beta^+$ $\beta^+$ , $\gamma$	81 sec 24.6 min	1.8, 3.3 ( $< 5\%$ ) abs. Al	1.5 abs. Pb	Ni-p-n Ni <sup>69</sup> -p-n Ni <sup>70</sup> -d-2n Ni <sup>68</sup> - $\alpha$ -p-n Cu-d-p4n As-d-5s17a
Cu <sup>71</sup>	B		$\beta^+$ ; K	3.4 hr; 3.33 hr	1,205 spect.; 0.9 abs.; 1.23	No $\gamma$	Ni-d-n Ni-p-n Ni <sup>71</sup> -p-n Ni-p- $\gamma$ Ni- $\alpha$ -p Cu- $\gamma$ -2n Cu-d-p3n or Cu-d-p5n As-d-5s16a
Cu <sup>72</sup>	A		$\beta^+$ , $\gamma$	10.5 min; 10.1 min	2.6 cl.ch.; 2.5 abs. Al	0.56 abs. Pb	Co- $\alpha$ -n Ni-p-n Ni-p- $\gamma$ Cu-n-2n Cu- $\gamma$ -n Cu-e-e-n Cu-d-i Zn <sup>62</sup> K-decay
Cu <sup>67</sup>		69.09					

Isotope Z      A	Class	Per cent abundance	Type of radiation	Half-life	Energy of radiation, Mev		Produced by
					Particles	$\gamma$ -rays	
29 Cu <sup>64</sup>	A		K(54 %); $\beta^-$ (31 %); $\beta^+$ (15 %); $\gamma$ (1.5 %) (with K)	12.8 hr	0.571( $\beta^-$ ), 0.657( $\beta^+$ ) spect.; 0.58( $\beta^-$ ), 0.66( $\beta^+$ ) spect.; 0.57( $\beta^-$ ), 0.64( $\beta^+$ ) spect.	1.35(2.5 %) spect.; 1.34(weak) spect.; 1.20(weak) coincid. abs.	Ni- $p-n$ Cu- $d-p$ Cu- $n-\gamma$ Cu- $n-2n$ Cu- $p-pn$ Cu- $\gamma-n$ Zn- $d-\alpha$ Zn- $n-p$ As- $d-5s13a$
Cu <sup>65</sup> Cu <sup>66</sup>	A	30.91	$\beta^-, \gamma$	5 min	2.9 cl.ch. (K.U.); 2.58	1.32 abs. Pb	Cu- $n-\gamma$ Cu- $d-p$ Zn- $n-p$ Ga- $n-\alpha$ Ni <sup>66</sup> $\beta^-$ -decay As- $d-5s10a$ Bi- $d$ Zn- $\gamma-p$
Cu <sup>67</sup>	B		$\beta^-$	56 hr; 61 hr	0.56 abs. Al		Cu- $d-3n$ or Cu- $d-5n$ , parent of Cu <sup>67</sup> As- $d-4s15a$
30 Zn <sup>62</sup>	A		K(?)	9.5 hr			Ni- $\alpha-n$ Cu- $p-n$ Cu- $d-2n$ Cu- $d-4n$ Zn- $n-2n$ Zn- $\gamma-n$ As- $d-4s14a$
Zn <sup>63</sup>	A		$\beta^+$ (93 %); K(7 %), $\gamma$	38 min	2.3 abs., spect.; 2.36(85 %), 1.40(7 %), 0.47(1 %) spect.	0.96(weak), 1.9(weak), 2.6(weak)	Cu- $d-2n$ Cu- $p-n$ Zn- $d-p$ Ga <sup>65</sup> K-decay
Zn <sup>64</sup> Zn <sup>65</sup>	A	48.89	$\beta^+$ (1.3 %), K(98.7 %), $\gamma, e^-$	250 days	0.32( $\beta^+$ ) spect.; 0.4( $\beta^+$ ) cl.ch.	1.11 spect.; 1.14 spect.; 1.14(46 % of K), no $\gamma$ (54 % of K) x-ray- $e^-$ coincid.; 0.45, 0.65, 1.0 cl.ch. recoil	Cu- $d-2n$ Cu- $p-n$ Zn- $d-p$ Zn- $n-\gamma$ Ga- $n-\alpha$ K-decay
Zn <sup>66</sup> Zn <sup>67</sup> Zn <sup>68</sup> Zn <sup>69m</sup>	A	27.81 4.07 18.61	I.T., $\gamma$	13.8 hr		0.439 spect. conv.	Zn- $d-p$ Zn- $n-\gamma$ Ga- $d-\alpha$ Ga- $n-p$ As- $d-2a$

Isotope Z      A	Class	Per cent abundance	Type of radiation	Half-life	Energy of radiation, Mev		Produced by
					Particles	$\gamma$ -rays	
30 Zn <sup>69</sup>	A		$\beta^-$	57 min	1.0 abs.	No $\gamma$	Zn-d- $\beta$ Zn-n- $\gamma$ Ga-d- $\alpha$ Ga-n- $\beta$ As-d-2 $\alpha$ Zn <sup>69m</sup> I.T.
Zn <sup>70</sup>	B	0.620	$\beta^-, \gamma$	2.2 min	2.1		Zn-n- $\gamma$ Ge-n- $\alpha$
Zn <sup>71</sup>	A		$\beta^-, \gamma$	49 hr	$\sim 0.3$ (95 %), $\sim 1.6$ (5 %) abs. Al		U-n, parent of Ga <sup>72</sup> Bi-d As-d-4z5a
31 Ga <sup>64</sup>	B		$\beta^+$	48 min			Zn-p-n
Ga <sup>65</sup>	A		K, $e^-$	15 min		0.054, 0.117 spect. conv.	Zn-d-n Zn-p- $\gamma$
Ga <sup>66</sup>	A		$\beta^+$	9.4 hr	3.1 abs.		Cu- $\alpha$ -n Zn-p-n As-d-3z11a Ge <sup>66</sup> decay
Ga <sup>67</sup>	A		K, $\gamma, e^-$	78.3 hr; 83 hr		0.094, 0.174, 0.187, 0.301 spect.; 0.0925, 0.180, 0.297 spect. conv., spect.; 0.292 spect.	Zn-d-n Zn- $\alpha$ - $\beta$ Zn-p-n As-d-3z10a Ge <sup>67</sup> $\beta^+$ -decay
Ga <sup>68</sup>	A		$\beta^+$	68 min	1.9 abs.		Cu- $\alpha$ -n Zn-p-n Zn-p- $\gamma$ (?) Zn-d-n Ga-n-2n Ga- $\gamma$ -n Ge- $\gamma$ -pn Ge-d- $\alpha$ As-d-3z9a Ge <sup>68</sup> K-decay
Ga <sup>69</sup> Ga <sup>70</sup>	A	60.2	$\beta^-, \gamma$	20.3 min; 20 min	1.68 cl.ch. (K.U.); 1.65 spect.; 1.62 abs. Al		Zn-p-n Zn- $\alpha$ - $\beta$ Ga-n- $\gamma$ Ga-n-2n Ga- $\gamma$ -n Ge-d- $\alpha$ Ge-n- $\beta$

Isotope Z      A	Class	Per cent abundance	Type of radiation	Half-life	Energy of radiation, Mev		Produced by
					Particles	$\gamma$ -rays	
31 Ga <sup>71</sup> Ga <sup>72</sup>	A	39.8	$\beta^-$ , $\gamma$	14.3 hr; 14.1 hr	0.64(40 %), 0.955 (32 %), 1.48 (10.5 %), 2.52(8 %), 3.15 (9.5 %) spect.; ~ 0.77, 2.3 co- incid. abs.; spect.; 0.8 (~ 65 %), ~ 3.1 (~ 35 %) abs. Al	0.63(24 %), 0.84 (100 %), 1.05 (4.5 %), 1.59 (4.5 %), 1.87 (7.8 %), 2.21 (33 %), 2.51 (26.5 %) spect.; 0.64 (~ 8 %), 0.84 (~ 46 %), 2.25 (~ 46 %) spect.; 2.50 D- $\gamma$ - $n$ reaction; spect.	Ga-d-p Ga-n- $\gamma$ Ge-n-p As-d- $\alpha$ p U-n, Zn <sup>72</sup> $\beta^-$ -decay Bi- $\alpha$ Tl- $\alpha$ U- $\alpha$
Ga <sup>72</sup>	B		$\beta^-$	5 hr	1.4 abs. Al	No $\gamma$	Ge-n-p Ge- $\gamma$ -p U-n
32 Ge <sup>66</sup>	A			~ 140 min			Ge-d-p5n, parent of Ga <sup>66</sup>
Ge <sup>67</sup>	A		$\beta^+$	23 min			Ge-d-p4n, parent of Ga <sup>67</sup>
Ge <sup>68</sup>	A		K	250 days; ~ 195 days			Zn- $\alpha$ -2n As-d- $\alpha$ 5n, parent of Ga <sup>68</sup>
Ge <sup>70</sup> Ge <sup>71</sup>	A	20.55	K, $e^-$ (?); K, no $\beta^-$ or $e^-$ ; $\beta^+$ (?)	11 days; 11.3 days; 11.4 days	~ 0.6( $\beta^+$ ?)	0.6 abs. of $e^-$	Ga-d-2n Ga-p-n Ge-d-p Ge-n- $\gamma$ As-d- $\alpha$ 2n Zn- $\alpha$ -n Ga-d-2n Ga-p-n Ge-n- $\gamma$ Ge-d-p Ge-n-2n Ge- $\gamma$ -n As-d- $\alpha$ 2n
Ge <sup>71</sup>	B		$\beta^+$	39.7 hr; 40 hr; 36 hr; 38 hr	1.2 abs.		

Isotope Z      A	Class	Per cent abundance	Type of radiation	Half-life	Energy of radiation, Mev		Produced by
					Particles	$\gamma$ -rays	
32 Ge <sup>71</sup>							Se-n- $\alpha$ As <sup>71</sup> $\beta^+$ - decay (?) Ga <sup>72</sup> $\beta^-$ -decay
Ge <sup>72m</sup>	A		I.T., $e^-$	$5 \times 10^{-7}$ sec	0.68( $e^-$ ) co- incid. abs.		
Ge <sup>72</sup>		27.37					
Ge <sup>73</sup>		7.61					
Ge <sup>74</sup>		36.74					
Ge <sup>75</sup>	A		$\beta^-, \gamma$	89 min	1.1 cl.ch. (K.U.); 1.2 abs. Al		Ge-n- $\gamma$ Ge-d-p Ge-n-2n Ge- $\gamma$ -n As-n-p Se-n- $\alpha$
Ge <sup>76</sup>		7.67					
Ge <sup>77</sup>	A		$\beta^-, \gamma$	12 hr	2.0 abs. Al; 1.9 cl.ch. (K.U.); 1.8 abs. Al		Ge-n- $\gamma$ Ge-d-p Se-n- $\alpha$ U-n, parent of As <sup>77</sup> U <sup>232</sup> -n Ge-n- $\gamma$ , parent of As <sup>77</sup>
Ge <sup>77m</sup>	B		$\beta^-$	59 sec	2.8 abs. Al		U-n, parent of As <sup>78</sup>
Ge <sup>78</sup>	D		$\beta^-, \gamma$	2.1 hr	$\sim 0.9$ abs. Al		As-d-p5n Se <sup>71</sup> $\beta^+$ -decay, parent of Ge <sup>71</sup> (11d.)
33 As <sup>71</sup>	B		$\beta^+$	52 min			Ga- $\alpha$ -n Ge-p-n As-d-p4n Se <sup>74</sup> -d- $\alpha$ Se <sup>72</sup> K-decay Ge-d-n Ge <sup>70</sup> - $\alpha$ -p (?)
As <sup>71</sup>	A		K	60 hr			
As <sup>72</sup>	B		$\beta^+, \gamma$	26 hr	2.78 abs. Al, coincid.	2.4 coincid. abs.	
As <sup>73</sup>	B		K, $e^-$	90 days		0.052 spect. conv.	
As <sup>74</sup>	A		$\beta^-, \beta^+, \gamma$	17.5 days; 19.0 days; 16 days	1.3( $\beta^-$ ), 0.9( $\beta^+$ ) cl.ch. (K.U.)	0.582 spect.	Ga- $\alpha$ -n As-n-2n As-d-p2n Ge-d-n Se-d- $\alpha$ Ge-p-n Bi-d
As <sup>75</sup>		100					
As <sup>76</sup>	A		$\beta^-, \gamma$ ; no $\beta^+$ ; $\beta^+$ , K, $\gamma$	26.8 hr	1.29 (15 %), 2.49 (25 %), 3.04 (60 %) ( $\beta^-$ ) spect.;	0.55, 1.20, 1.70 spect.; 0.557, 1.22, 1.78	Ge-p-n As-d-p As-n- $\gamma$ Se-n-p Se- $\gamma$ -p



Isotope Z      A	Class	Per cent abundance	Type of radiation	Half-life	Energy of radiation, Mev		Produced by
					Particles	$\gamma$ -rays	
33 As <sup>76</sup>					1.1, 1.7, 2.7( $\beta^-$ ) cl.ch.; 0.7, 2.6( $\beta^+$ ) cl.ch.; coincid.	(weak) spect.; 1.94, 0.83 spect.; coincid.; 2.15 (weak), 1.84 (weak), 1.25 ( $\sim 30\%$ ), 0.57 ( $\sim 70\%$ ) spect.; 3.2, 2.2, 1.5 cl.ch. pair	Se-d- $\alpha$ Br-n- $\alpha$
As <sup>77</sup>	A		$\beta^-$	40 hr	0.8 abs. Al		U-n, Ge <sup>77</sup> $\beta^-$ -decay Th- $\alpha$ Bi-d Ge <sup>77</sup> (59 sec) $\beta^-$ -decay
As <sup>78</sup>	A		$\beta^-, \gamma$	80 min; 65 min	1.4 cl.ch. (K.U.)	0.27 abs. Pb	Br-n- $\alpha$ Se-n-p U-n, Ge <sup>78</sup> $\beta^-$ -decay
As <sup>78</sup>	D		$\beta^-$	90 min	1.4 ( $\sim 30\%$ ), 4.1 ( $\sim 70\%$ ) abs. Al		
34 Se <sup>71</sup>	B		$\beta^+$	44 min			As-d-6n, parent of As <sup>71</sup>
Se <sup>72</sup>	B		K	9.5 days			As-d-5n, parent of As <sup>72</sup>
Se <sup>72</sup>	B		$\beta^+; K$	6.7 hr 7.1 hr	1.29 abs. Al		Ge- $\alpha$ -n Ge <sup>70</sup> - $\alpha$ -n As-d-4n
Se <sup>74</sup> Se <sup>75</sup>	A	0.87	K, $\gamma$ , $e^-$	127 days; 125 days; 115 days; 120 days		0.077, 0.099, 0.124, 0.139, 0.269, 0.281, 0.405 spect., spect. conv.; 0.50 spect. conv.; several	As-p-n As-d-2n Se-n- $\gamma$

Isotope Z      A	Class	Per cent abundance	Type of radiation	Half-life	Energy of radiation, Mev		Produced by
					Particles	$\gamma$ -rays	
34 Se <sup>75</sup>						< 0.3 spect. conv.; 0.335, 0.18; 0.22, 0.43 abs. Pb	
Se <sup>76</sup> Se <sup>77</sup> Se <sup>77m</sup>	A	9.02 7.58	I.T., $\gamma$	17.5 sec	0.135(e <sup>-</sup> ) abs.	~ 0.15	Se-n- $\gamma$ Se <sup>76</sup> n- $\gamma$ Se-x-rays
Se <sup>78</sup> Se <sup>80</sup> Se <sup>81m</sup>	B	23.52 49.82	I.T., e <sup>-</sup>	59 min; 57 min		0.099 spect. conv.	Se-d- $\beta$ Se-n- $\gamma$ Se <sup>80</sup> n- $\gamma$ Se- $\gamma$ -n Br-n- $\beta$ U-n, parent of Se <sup>81</sup> Se-d- $\beta$ Se-n- $\gamma$ Se- $\gamma$ -n Se <sup>81m</sup> I.T. Br-n- $\beta$ U-n, Se <sup>81m</sup> I.T.
Se <sup>81</sup>	B		$\beta^-$	17 min; 19 min	1.5 abs. Al	No $\gamma$	Se-d- $\beta$ Se-n- $\gamma$ Se- $\gamma$ -n Se <sup>81m</sup> I.T. Br-n- $\beta$ U-n, Se <sup>81m</sup> I.T.
Se <sup>82</sup> Se <sup>83m</sup>	A	9.19	$\beta^-$ , $\gamma$	67 sec	3.4 abs. Al		Se-n- $\gamma$ U-n
Se <sup>83</sup>	A		$\beta^-$ , $\gamma$	25 min; 30 min	1.5 abs. Al	0.17, 0.37, 1.1 abs. Pb	Se-d- $\beta$ Se-n- $\gamma$ U-n, parent of Br <sup>83</sup> Th-n
Se <sup>84</sup>	A		$\beta^-$	~ 2.5 min; < 10 min			U-n, parent of Br <sup>84</sup>
35 Br <sup>75</sup>	B		$\beta^+$ ; K	1.7 hr	1.6 abs. Al	No $\gamma$	Se <sup>74</sup> d-n Se <sup>74</sup> p- $\gamma$ (?), parent of Se <sup>75</sup> (?) As- $\alpha$ -3n
Br <sup>76</sup>	D		$\beta^+$ , $\gamma$ e <sup>-</sup>	15.7 hr	3.15( $\beta^+$ ), 0.18(e <sup>-</sup> ) spect.	2 abs. Pb	
Br <sup>77</sup>	B		$\beta^+$ ; K, $\gamma$ , e <sup>-</sup> ; K(95 %), $\beta^+$ (5 %)	57.2 hr; 58 hr	0.36( $\beta^+$ ) spect., abs. Al, spect.	0.7 abs. Pb	As- $\alpha$ -2n Se <sup>74</sup> $\alpha$ - $\beta$ Se <sup>76</sup> d-n
Br <sup>78</sup>	A		$\beta^+$ , e <sup>-</sup> , $\gamma$	6.4 min	2.3( $\beta^+$ ) abs.	0.046, 0.108 spect. conv.	As- $\alpha$ -n Se-d-n Se-p-n Br- $\gamma$ -n Br-n-2n

Isotope $Z$ $A$	Class	Per cent abundance	Type of radiation	Half-life	Energy of radiation, Mev		Produced by
					Particles	$\gamma$ -rays	
35 Br <sup>79</sup> Br <sup>80m</sup>	A	50.5	I.T., $e^-$ , $\gamma$	4.4 hr		0.049, 0.037, or 0.025 spect. conv.; 0.037 abs. Al	Se- $\alpha$ - $p$ Se- $p$ - $n$ Br- $n$ - $\gamma$ , ( $\sim 30\%$ ) Br- $d$ - $p$ Br- $\gamma$ - $n$ Br- $n$ -2 $n$ Th- $n$ (?) Se- $p$ - $n$ Br- $n$ - $\gamma$ , ( $\sim 70\%$ ) Br- $d$ - $p$ Br- $\gamma$ - $n$ Br- $n$ -2 $n$ Br <sup>80m</sup> I.T.
Br <sup>80</sup>	A		$\beta^-$ , $\gamma$ ; $\beta^+$ (3%)	18 min	2.0( $\beta^-$ ) spect.; 0.73( $\beta^+$ ) spect., abs.	< 0.5 abs.	
Br <sup>81</sup> Br <sup>82</sup>	A	49.5	$\beta^-$ , $\gamma$	34 hr	0.465 spect.; coincid.	0.547, 0.787, 1.35 spect.; coincid.	Se- $p$ - $n$ Se- $d$ -2 $n$ Br- $n$ - $\gamma$ Br- $d$ - $p$ Rb- $n$ - $\alpha$ U- $n$ Pb- $\alpha$ Tl- $\alpha$ Bi- $\alpha$ Bi- $d$ U- $\alpha$
Br <sup>83</sup>	A		$\beta^-$	2.4 hr; 140 min	1.05 abs.; 0.9 abs. Al	No $\gamma$	Se- $d$ - $n$ Se <sup>83</sup> $\beta^-$ -de- cay, parent of Kr <sup>83m</sup> U- $n$ , Se <sup>83</sup> $\beta^-$ -decay, parent of Kr <sup>83m</sup> U <sup>232</sup> - $n$ Th- $n$ Th- $\alpha$ Pu- $n$ Bi- $d$ Pb- $\alpha$ Bi- $\alpha$ U- $\alpha$
Br <sup>84</sup>	A		$\beta^-$ , $\gamma$	30 min; 33 min	5.3 abs. Al; 4.5 abs.		Rb- $n$ - $\alpha$ U- $n$ , Se <sup>84</sup> $\beta^-$ -decay Th- $n$ Bi- $d$
Br <sup>85</sup>	A		$\beta^-$	3.00 min; 3.0 min	2.5 abs. Al	No $\gamma$	U- $n$ , parent of Kr <sup>85</sup>
Br <sup>87</sup>	B		$\beta^-$ ; $\beta$ , $n$ $\beta^-$	55.6 sec; 55.0 sec; 56 sec	0.25 (mean) ( $n$ ) abs. paraffin;		U- $n$ , parent of Kr <sup>87</sup> Pu- $n$

Isotope <i>Z A</i>	Class	Per cent abundance	Type of radiation	Half-life	Energy of radiation, Mev		Produced by
					Particles	$\gamma$ -rays	
35 Br <sup>87</sup>							
Br <sup>87</sup>	D		$\beta^-$ , $n$	4.51 sec; 4.5 sec	0.3 (mean) ( $n$ ) $p$ re- coil in cl.ch. 0.43 (mean) ( $n$ ) abs. paraffin; 0.7 (mean) ( $n$ ) $p$ re- coil in cl.ch.		U- $n$
Br <sup>88</sup>	B		$\beta^-$	16.0 sec			U- $n$ , ances- tor of Rb <sup>88</sup>
36 Kr <sup>77</sup>	B		K (70 %), $\beta^+$ (30 %), $\gamma$	1.1 hr	1.7 abs. Al		Se- $\alpha$ - $n$ Se <sup>74</sup> - $\alpha$ - $n$
Kr <sup>78</sup>	A	0.342	$\beta^+$ , (2 %); $\gamma$ ; K (98 %)	34 hr	$\sim 0.9$ (30 %), $\sim 0.6$ (70 %) abs. Al; 0.4 cl.ch.	0.2 abs. Pb	Se- $\alpha$ - $n$ Se <sup>78</sup> - $\alpha$ - $n$ Br- $d$ -2 $n$ Br- $p$ - $n$ Kr- $d$ - $p$ Kr- $n$ - $\gamma$ Br- $p$ - $n$
Kr <sup>79,81</sup>	C		I.T.(?), $e^-$ , $\gamma$ ; no $\beta^+$	13 sec		0.187 spect. conv.	Se- $\alpha$ - $n$ (?) Br- $p$ - $n$
Kr <sup>79,81</sup>	C		I.T.(?), $e^-$ , $\gamma$ ; no $\beta^+$	55 sec		0.127 spect. conv.	
Kr <sup>80</sup>		2.223					
Kr <sup>82</sup>		11.50					
Kr <sup>83</sup>		11.48					
Kr <sup>83m</sup>	A		I.T., $e^-$	113 min		0.029, 0.046 spect. conv.	U- $n$ m.s. Se- $\alpha$ - $n$ Kr- $d$ - $p$ Kr- $n$ - $\gamma$ Kr- $x$ -rays U- $n$ , Br <sup>88</sup> $\beta^-$ -decay
Kr <sup>84</sup>		57.02					U- $n$ m.s.
Kr <sup>85</sup>	A		$\beta^-$ , $\gamma$	4.5 hr; 4.0 hr; 4.6 hr	1.0 abs. Al; 0.85 abs	0.17, 0.37 abs. Pb	Kr- $d$ - $p$ Kr- $n$ - $\gamma$ Rb- $n$ - $p$ Sr- $n$ - $\alpha$ U- $n$ , Br <sup>88</sup> $\beta^-$ -decay
Kr <sup>86</sup>	B		$\beta^-$	9.4 yr; $\sim 10$ yr; > 2.5 yr	0.74 abs. Al; $\sim 0.8$ abs. Al	No $\gamma$	Kr- $n$ - $\gamma$ U- $n$
Kr <sup>86</sup>	m.s.						
Kr <sup>86</sup>		17.43					
Kr <sup>87</sup>	B		$\beta^-$	74 min	$\sim 4$ abs. Al		U- $n$ m.s. Kr- $d$ - $p$ Rb- $n$ - $p$ U- $n$ , Br <sup>87</sup> $\beta^-$ -decay
Kr <sup>88</sup>	A		$\beta^-$	3 hr	2.5 cl.ch. (K.U.)		Th- $n$ U- $n$ , parent of Rb <sup>88</sup>

Isotope Z A	Class	Per cent abundance	Type of radiation	Half-life	Energy of radiation, Mev		Produced by
					Particles	$\gamma$ -rays	
36 Kr <sup>82</sup>	A		$\beta^-$	2.6 min; 2.5 min			U- $n$ , ances- tor of Sr <sup>80</sup>
Kr <sup>90</sup>	A		$\beta^-$	$\sim 33$ sec; short			U- $d$ Pu- $n$ U- $n$ , ances- tor of Sr <sup>90</sup>
Kr <sup>91</sup>	B		$\beta^-$	9.3 sec; 5.7 sec			Pu- $n$ U- $n$ , ances- tor of Sr <sup>91</sup> , ancestor of Y <sup>91</sup>
Kr <sup>92</sup>	A		$\beta^-$	2.3 sec; < 0.5 min			U- $d$ Pu- $n$ U- $n$ , ances- tor of Y <sup>92</sup>
Kr <sup>93</sup>	A		$\beta^-$	2.2 sec; 2.0 sec			Th- $n$ Pu- $n$ U- $n$ , ances- tor of Y <sup>93</sup>
Kr <sup>94</sup>	B		$\beta^-$	1.4 sec			U- $d$ Pu- $n$ U- $n$ , ances- tor of Y <sup>94</sup>
Kr <sup>97</sup>	B		$\beta^-$	Short			U- $n$ , ances- tor of Zr <sup>97</sup> Pu- $n$
37 Rb <sup>81</sup>	A m.s.		$\beta^+$ , $\gamma$ , $e^-$	5.0 hr	0.9( $\beta^+$ ), 0.2( $e^-$ ) abs. Al, spect.	0.8 abs. Pb	Br- $\alpha$ -2 $n$
Rb <sup>82</sup>	A m.s.		$\beta^+$ , $\gamma$	6.3 hr; 6.5 hr	0.9 abs. Al	1.0 abs. Pb	Br- $\alpha$ - $n$ Kr- $d$ -2 $n$
Rb <sup>83</sup>	D			20 min			Br- $\alpha$ - $n$
Rb <sup>84</sup>	B		$\beta^+$	$\sim 40$ days			Rb- $n$ -2 $n$ Sr- $d$ - $\alpha$
Rb <sup>85</sup>		72.8					
Rb	F			42 min			Kr- $d$ - $n$
Rb	F			200 hr			Kr- $d$ - $n$
Rb <sup>86</sup>	A		$\beta^-$ , $\gamma$	19.5 days; $\sim 17$ days	1.82(80 %), 0.716 (20 %) spect., coincid.; coincid.; coincid., 1.56 abs.; 1.60 spect.; 1.80 abs. Al	1.081 spect., coincid.; coincid.	Rb- $n$ - $\gamma$ Rb- $\gamma$ - $n$ Sr- $d$ - $\alpha$ Bi- $d$ U- $n$
Rb <sup>87</sup>	A	27.2	$\beta^-$ , $\gamma$ , $e^-$	$6.3 \times 10^{10}$ yr; $5.8 \times 10^{10}$ yr;	0.132 spect.; 0.25; 0.13	0.034, 0.053, 0.082, 0.102,	Natural source

Isotope $Z$ $A$	Class	Per cent abundance	Type of radiation	Half-life	Energy of radiation, Mev		Produced by
					Particles	$\gamma$ -rays	
37 Rb <sup>87</sup>							
Rb <sup>88</sup>	A		$\beta^-$	$1.2 \times 10^{11}$ yr 17.5 min	spect.; 0.144 spect. 4.6 abs. Al; 5.1 cl.ch.	0.129 spect. conv.	Rb- $n$ - $\gamma$ Pa- $n$ U- $n$ , Kr <sup>88</sup> $\beta^-$ -decay Th- $n$ U- $n$ , Kr <sup>89</sup> $\beta^-$ -decay; parent of Sr <sup>89</sup>
Rb <sup>89</sup>	A		$\beta^-$ , $\gamma$	15 min	3.8 abs.		U- $n$ , Kr <sup>90</sup> $\beta^-$ -decay, parent of Sr <sup>90</sup>
Rb <sup>90</sup>	A		$\beta^-$	Short			U- $n$ , Kr <sup>91</sup> $\beta^-$ -decay, ancestor of Y <sup>91</sup>
Rb <sup>91</sup>	A		$\beta^-$	Short			U- $n$ , Kr <sup>92</sup> $\beta^-$ -decay, ancestor of Y <sup>92</sup>
Rb <sup>&gt;90</sup>	D		$\beta^-$	80 sec			U- $n$
Rb <sup>93</sup>	A		$\beta^-$	Short			U- $n$ , Kr <sup>93</sup> $\beta^-$ -decay, ancestor of Y <sup>93</sup>
Rb <sup>94</sup>	B		$\beta^-$	Short			U- $n$ , Kr <sup>94</sup> $\beta^-$ -decay, ancestor of Y <sup>94</sup>
Rb <sup>97</sup>	B		$\beta^-$	Short			U- $n$ , Kr <sup>97</sup> $\beta^-$ -decay, ancestor of Zr <sup>97</sup>
38 Sr <sup>84</sup>							
Sr <sup>86m</sup>	A	0.56	I.T., $e^-$ , $\gamma$	70 min		0.170 spect. conv.	Rb- $p$ - $n$
Sr <sup>86</sup>	A		K, $\gamma$	65 days		0.8 abs. Pb	Rb- $p$ - $n$ Rb- $d$ -2 $n$
Sr <sup>86</sup>		9.86					
Sr <sup>87m</sup>	A		I.T., $e^-$ , $\gamma$	2.7 hr		0.37 spect. conv.; 0.386 spect. conv.	Rb- $p$ - $n$ Sr- $n$ - $n$ Sr- $x$ -rays Sr- $e^-$ - $e^-$ Sr- $d$ - $p$ Sr- $n$ - $\gamma$ Sr <sup>86</sup> - $n$ - $\gamma$ Sr- $p$ - $p$ (?) Y <sup>87</sup> K-decay Zr- $n$ - $\alpha$
Sr <sup>87</sup>		7.02					
Sr <sup>88</sup>		82.56					
Sr <sup>89</sup>	A m.s.		$\beta^-$	53 days; 55 days	1.50 cl.ch.; 1.48	No $\gamma$	Sr- $d$ - $p$ Sr- $n$ - $\gamma$

Isotope Z     A	Class	Per cent abundance	Type of radiation	Half-life	Energy of radiation, Mev		Produced by
					Particles	$\gamma$ -rays	
38 Sr <sup>86</sup>					spect.; 1.5 spect.		Y-n-p Zr-n- $\alpha$ (?) U-n, Rb <sup>86</sup> $\beta$ -decay U-d U <sup>232</sup> -n Th-n Th- $\alpha$ Pu-n Bi- $\alpha$ Bi-d Pb- $\alpha$ Pt- $\alpha$
Sr <sup>90</sup>	A m.s.		$\beta^-$	25 yr; ~ 30 yr	0.61 spect.; 0.6 abs. Al	No $\gamma$	U-n, Rb <sup>90</sup> $\beta$ -decay, parent of Y <sup>90</sup> U <sup>232</sup> -n Th- $\alpha$
Sr <sup>91</sup>	A		$\beta^-, \gamma$	9.7 hr; 10 hr	1.3 (40 %). 3.2 (60 %) abs. Al	~ 1.3 abs. Pb	Zr-n- $\alpha$ U-n, Rb <sup>91</sup> $\beta$ -decay, parent of Y <sup>91</sup> (~ 60 %) and Y <sup>91m</sup> (~ 40 %) Th-n Th- $\alpha$ Pu-n Bi- $\alpha$ Pt- $\alpha$ Pb- $\alpha$ Bi-d
Sr <sup>92</sup>	A		$\beta^-$	2.7 hr			U-n, parent of Y <sup>92</sup> Th-n Th- $\alpha$ U- $\gamma$
Sr <sup>93</sup>	A		$\beta^-$	7 min			U-n, Rb <sup>93</sup> $\beta$ -decay, parent of Y <sup>93</sup>
Sr <sup>94</sup>	B		$\beta^-$	~ 2 min			U-n, Rb <sup>94</sup> $\beta$ -decay, parent of Y <sup>94</sup>
Sr <sup>97</sup>	B		$\beta^-$	Short			U-n, Rb <sup>97</sup> $\beta$ -decay, ancestor of Zr <sup>97</sup>
39 Y <sup>87m</sup>	B		I.T., e <sup>-</sup> , $\gamma$	14 hr		0.5 abs.	Sr-d-n Sr-p-n

Isotope $Z \quad A$	Class	Per cent abundance	Type of radiation	Half-life	Energy of radiation, Mev		Produced by
					Particles	$\gamma$ -rays	
39 Y <sup>87</sup>	A		K	80 hr		No $\gamma$ (?)	Rb- $\alpha$ - $n$ Sr- $p$ - $n$ Sr- $d$ - $n$ Sr- $d$ - $n$ Sr- $p$ - $n$ Y- $n$ - $2n$
Y <sup>88</sup>	A		$\beta^+$	2.0 hr	1.65 abs. Al; 1.2 cl.ch. (K.U.)		
Y <sup>88</sup>	A m.s.		K, $\gamma$ ; $\beta^+$ (0.19 %)	105 days	0.83 ( $\beta^+$ ) spect.	0.908, 1.853, 2.76 spect; 0.908, 1.89 spect. coincid.; 0.95, 1.92 cl.ch.; 1.87 Be- $\gamma$ - $n$ ; 2.8 (1 %) D- $\gamma$ - $n$	Sr- $p$ - $n$ Sr- $d$ - $2n$ Y- $n$ - $2n$
Y <sup>90</sup>		100					
Y <sup>90</sup>	A m.s.		$\beta^-$	62 hr; 65 hr 60 hr	2.35 spect.; 2.16 spect.; 2.6 cl.ch. (K.U.); 2.5 abs. Al	No $\gamma$	Y- $d$ - $p$ Y- $n$ - $\gamma$ Zr- $n$ - $p$ Zr- $d$ - $\alpha$ Cb- $n$ - $\alpha$ U- $n$ , Sr <sup>90</sup> $\beta^-$ -decay Bi- $d$ Bi- $\alpha$ Pt- $\alpha$ Tl- $\alpha$ Zr- $n$ - $p$ U- $n$ , Sr <sup>91</sup> $\beta^-$ -decay Zr- $n$ - $p$ U- $n$ , Sr <sup>91</sup> $\beta^-$ -decay; Y <sup>91m</sup> I.T. U <sup>232</sup> - $n$ U- $d$ Th- $n$ Pu- $n$ Bi- $d$ Zr- $n$ - $p$ U- $n$ , Sr <sup>91</sup> $\beta^-$ -decay Th- $n$ Pu- $n$ U- $n$ , Sr <sup>91</sup> $\beta^-$ -decay Th- $n$ Pu- $n$ Zr- $n$ - $p$ U- $n$ , Sr <sup>94</sup> $\beta^-$ -decay Pu- $n$
Y <sup>91m</sup>	A		I.T., $\gamma$ , $e^-$ (~9 %)	51.0 min; 50 min		0.61 abs. Pb, abs. Al of $e^-$	
Y <sup>91</sup>	A m.s.		$\beta^-$	57 days; 61 days	1.53 spect.; 1.6 abs.	No $\gamma$	
Y <sup>92</sup>	A		$\beta^-, \gamma$	3.5 hr	3.5 abs. Al; 3.6 abs. Al	~1 abs Pb	
Y <sup>92</sup>	A		$\beta^-, \gamma$	10.0 hr; 11.5 hr	3.1 abs. Al	0.7 abs. Pb	
Y <sup>94</sup>	B		$\beta^-, \gamma$	20 min			



Isotope Z      A	Class	Per cent abundance	Type of radiation	Half-life	Energy of radiation, Mev		Produced by
					Particles	$\gamma$ -rays	
39 Y <sup>97</sup>	B		$\beta^-$	Short			U-n, Sr <sup>97</sup> $\beta^-$ -decay, parent of Zr <sup>97</sup>
40 Zr <sup>88</sup>	A		$e^-$ , $\gamma$ , I.T. or K	4.5 min			Y-p-n Zr-n-2n(?)
Zr <sup>89</sup>	A		$\beta^+$	80.1 hr; 78 hr	1.07 abs. Al; 1.0( $\beta^+$ ) cl.ch. (K.U.), abs.	No $\gamma$	Y-d-2n Y-p-n Zr-n-2n Mo-n- $\alpha$
Zr <sup>90</sup>		51.46					
Zr <sup>91</sup>		11.23					
Zr <sup>92</sup>		17.11					
Zr <sup>94</sup>		17.40					
Zr <sup>96</sup>	A		$\beta^-$ , $\gamma$ , $e^-$	65 days; 65.5 days; 63 days	0.394 (98 %), 1.0(2 %) spect.; 0.42 (95 %), 1.0(5 %) abs. Al	0.73(93 %), 0.23 (93 %), 0.92(7 %) spect. conv.; 0.80 abs. Pb	Zr-n- $\gamma$ Zr-d-p Mo-n- $\alpha$ U-n, parent of Cb <sup>96</sup> (35 days) and Cb <sup>96</sup> (90 hr)(?) U <sup>232</sup> -n Pu-n U- $\alpha$ Bi-d Th- $\alpha$
Zr <sup>98</sup>		2.80					
Zr <sup>97</sup>	B		$\beta^-$ , $\gamma$	17.0 hr	2.2 abs. Al; 1 abs.	$\sim$ 0.8 abs. Pb	Zr-n- $\gamma$ Mo-n- $\alpha$ U-n, Sr <sup>97</sup> $\beta^-$ -decay, parent of Cb <sup>97</sup> U- $\alpha$ Th- $\alpha$ Pu-n
Zr	E			5 sec			Zr-n- $\gamma$ (?)
Zr	E		$\beta^-$	18 min			Zr-n- $\gamma$ (?)
Zr	F		$\beta^-$	90 min	$\sim$ 1.5 abs.		Zr-d-?
Zr	E		$\beta^-$	70 hr	1.17 cl.ch. (K.U.)		Zr-n-?
41 Cb	E			4 min			Zr-p-n(?)
Cb	E			12 min			Zr-p-n(?)
Cb	E			38 min			Zr-p-n(?)
Cb <sup>96</sup>	B		$\beta^+$ , $\gamma$	15.6 hr; 18 hr; 21 hr	$\sim$ 1 abs. Al	1 abs. Pb	Zr-p-n(?) Zr-d-2n Mo <sup>92</sup> -d- $\alpha$ Mo-d- $\alpha$
Cb <sup>91m</sup>	A		I.T., $e^-$ , $\gamma$	62 days; 60 days; $\sim$ 55 days		$\sim$ 0.15 abs. of $e^-$ ; 0.94	Zr-d-n Mo <sup>94</sup> -d- $\alpha$

Isotope Z      A	Class	Per cent abundance	Type of radiation	Half-life	Energy of radiation, Mev		Produced by
					Particles	$\gamma$ -rays	
41 Cb <sup>92</sup>	A	100	$\beta^-$ , $\gamma$	10.1 days; 11 days	1.38 cl.ch. (K.U.); 1.38; 0.59	1.0	Zr- <i>p-n</i> Cb- <i>n-2n</i> Cb- <i>d-t</i> Mo- <i>n-p</i> Mo <sup>94</sup> - <i>d-<math>\alpha</math></i> Cb- <i>d-t</i> Mo <sup>94</sup> - <i>d-<math>\alpha</math></i>
Cb <sup>92</sup>	A		$\beta^-$ , $\gamma$	21.6 hr	1.2 abs. Al	0.6 abs. Pb	
Cb <sup>92</sup>	F		I.T.	42 days			Cb-x-rays
Cb <sup>92m</sup>	A		I.T., $e^-$ ( $\sim 99.9\%$ ), $\beta^-$ ( $\sim 0.1\%$ )	6.6 min	1.3 coincid. abs. Al	0.058 abs. of $e^-$ ; 1.0 abs. Pb	Cb- <i>n-<math>\gamma</math></i> Cb- <i>d-<math>\gamma</math></i>
Cb <sup>94</sup>	A			$> 10^4$ yr			Cb- <i>n-<math>\gamma</math></i> , Cb <sup>94m</sup> I.T.
Cb <sup>95m</sup>	A		I.T., $e^-$ (100%)	90 hr; 80 hr		0.216 spect.; 0.24 spect. conv.	U- <i>n</i> , Zr <sup>95</sup> $\beta^-$ -decay ( $\sim 2\%$ ), parent of Cb <sup>95</sup>
Cb <sup>95</sup>	A		$\beta^-$ , $\gamma$ , $e^-$	35 days; 37 days	0.146 spect.; 0.15 abs. Al; 0.154 spect.	0.75 spect.; 0.79 spect.; 0.775 spect. conv.; 0.92 co- incid. abs., coincid.	Mo <sup>97</sup> - <i>d-<math>\alpha</math></i> Zr <sup>95</sup> $\beta^-$ -decay Mo- <i>d-<math>\alpha</math></i> Mo <sup>97</sup> - <i>d-<math>\alpha</math></i> U- <i>n</i> , Zr <sup>95</sup> $\beta^-$ -decay ( $\sim 98\%$ )
Cb <sup>96</sup>	A		$\beta^-$ , $\gamma$	2.8 days; 3 days; 4 days	1.8 abs. Al	1 abs. Pb, coincid. abs.	Zr- <i>p-n</i> Zr- <i>d-2n</i> Mo- <i>d-<math>\alpha</math></i>
Cb <sup>97</sup>	A		$\beta^-$ , $\gamma$	68 min; 75 min	1.4 abs. Al	0.78 abs. Pb	Mo <sup>98</sup> - <i>d-<math>\alpha</math></i> Mo- <i>n-p</i> Mo- <i>γ-p</i> Mo <sup>100</sup> - <i>d-<math>\alpha</math></i> U- <i>n</i> , Zr <sup>97</sup> $\beta^-$ -decay Mo <sup>100</sup> - <i>d-<math>\alpha</math></i>
Cb <sup>98</sup>	A		$\beta^-$	30 min			
Mo <sup>92</sup>	B	15.86	$\beta^+$ , $\gamma$	6.70 hr; 7 hr	0.3, 0.7	1.6	Zr- $\alpha$ - <i>n</i> Cb- <i>p-n</i> Cb- <i>d-2n</i> Mo- <i>d-p</i> Cb- <i>d-2n</i> Mo- <i>n-2n</i> Mo- <i>γ-n</i> Mo- <i>d-p</i>
Mo <sup>93</sup>	F		$\beta^+$	17 min	2.65 cl.ch. (K.U.)		
Mo <sup>94</sup>		9.12					
Mo <sup>95</sup>		15.7					
Mo <sup>96</sup>		16.5					
Mo <sup>97</sup>		9.45					
Mo <sup>98</sup>		23.75					

Isotope Z A	Class	Per cent abundance	Type of radiation	Half-life	Energy of radiation, Mev		Produced by
					Particles	$\gamma$ -rays	
42 Mo <sup>99</sup>	A		$\beta^-$ , $\gamma$	67 hr; 66.0 hr	1.3 abs. Al; 1.5 abs.; 0.24, 1.03 coincid. abs.	0.4 abs. Cu, Pb; 0.24 (20 %), Mo <sup>98</sup> - $n$ - $\gamma$ Mo <sup>98</sup> - $n$ - $\gamma$ Mo <sup>98</sup> - $n$ - $\gamma$ spect.; 0.77, 0.815, 0.84 spect.; 0.71 co- incid abs.	Zr- $\alpha$ - $n$ Mo- $d$ - $p$ Mo- $n$ - $\gamma$ Mo <sup>98</sup> - $n$ - $\gamma$ Mo- $n$ - $2n$ U- $n$ , parent of Tc <sup>99m</sup> U <sup>232</sup> - $n$ Th- $n$ Th- $\alpha$ Pu- $n$ Bi- $\alpha$ Bi- $d$ Tl- $\alpha$ Pt- $\alpha$
Mo <sup>100</sup> Mo <sup>101</sup>	A	9.6	$\beta^-$ , $\gamma$	14.6 min	1.0, 2.2; 1.8 cl.ch. (K.U.)	0.3, 0.9	Mo- $n$ - $\gamma$ Mo <sup>100</sup> - $n$ - $\gamma$ U- $n$ , parent of Tc <sup>101</sup> U- $n$ , parent of Tc <sup>102</sup> U- $n$ , ances- tor of Ru <sup>106</sup>
Mo <sup>102</sup>	D		$\beta^-$	12 min			
Mo <sup>106</sup>	B		$\beta^-$	Short			
43 Tc <sup>92</sup> Tc <sup>92.52</sup>	B C		$\beta^+$ , $\gamma$ $\beta^+$ , $\gamma$	4.5 min 2.7 hr	4.3 abs. 1.2 abs.	1.3 abs. 2.4 abs. Pb	Mo <sup>92</sup> - $d$ - $2n$ Mo <sup>92</sup> - $d$ - $2n$ Mo- $p$ - $n$ Mo- $d$ - $n$ Mo- $p$ - $n$ Mo <sup>94</sup> - $d$ - $2n$
Tc <sup>94m</sup>	B		I.T., $e^-$	53 min		0.0334 spect. conv.	Mo- $p$ - $n$ Mo <sup>94</sup> - $d$ - $2n$
Tc <sup>94</sup>	B		$\beta^+$ ; K(65 %), $\gamma$	< 53 min	2.47( $\beta^+$ ) spect.; 2.5( $\beta^+$ ) abs. Al	0.380, 0.873, 1.48, 1.85, 2.74 spect.	Mo- $p$ - $n$ Mo <sup>94</sup> - $d$ - $2n$
Tc <sup>96</sup>	A		K, $\gamma$ , $e^-$ ; $\beta^+$ ( $\sim 1$ %)	56 days; 52 days; 62 days	0.4( $\beta^+$ ) cl.ch.	0.25, 0.84 abs. Pb; 0.201, 0.57, 0.81, 1.01 spect., spect. conv., coincid.	Mo- $d$ - $n$ Mo- $p$ - $n$ Mo <sup>96</sup> - $d$ - $2n$
Tc <sup>96</sup>	A		K, $\gamma$ , $e^-$	20.0 hr		0.762, 0.932, 1.071 spect. conv.; 0.78 abs. Pb; 0.8 abs. Pb	Mo- $p$ - $n$ Mo- $d$ - $n$ Mo <sup>96</sup> - $d$ - $2n$ Ru <sup>96</sup> $\beta^+$ -decay
Tc <sup>96</sup>	A		K, $e^-$ (?), $\gamma$	4.30 days; 4.33 days	0.64( $e^-$ ) abs. Al; no	0.312, 0.771,	Cb- $\alpha$ - $n$ Mo- $p$ - $n$

Isotope Z      A	Class	Per cent abundance	Type of radiation	Half-life	Energy of radiation, Mev		Produced by
					Particles	$\gamma$ -rays	
43 Tc <sup>96</sup>					$\beta^-$ , no $e^-$ , $\sim 0.8(\beta^-?)$ spect.	0.806, 0.842, 1.119 spect. conv., spect., coincid.; 0.92 spect.; 0.8 abs. Pb	Mo- $d-n$ Ru- $n-p$ Mo <sup>96</sup> - $d-2n$
Tc <sup>97m</sup>	A		I.T., $e^-$	90 days; 93 days; 95 days		0.097 spect. conv.; 0.108 abs. of $e^-$	Mo <sup>97</sup> - $d-2n$ Mo- $d-n$ Mo- $p-n$ Ru <sup>97</sup> K-decay Mo <sup>97</sup> - $d-2n$ , Tc <sup>97m</sup> I.T.
Tc <sup>97</sup>	A			> 100 yr			Mo <sup>98</sup> - $d-2n$ Ru- $n-p$
Tc <sup>98</sup>	B		$\beta^-$ ; K(?), $\gamma$	2.7 days; 2.8 days	1.3 abs. Al; 0.75 abs. Al	0.9 abs. Pb; 1.0 abs. Pb	Mo <sup>98</sup> - $d-2n$ Ru- $n-p$
Tc <sup>99m</sup>	A		I.T., $e^-$ , $\gamma$	6.0 hr; 5.9 hr; 6.6 hr		0.136 spect. conv.; $\sim 0.18$ abs. Cu, Pb	Mo <sup>99</sup> $\beta^-$ -decay Ru- $n-p$ U- $n$ , Mo <sup>99</sup> $\beta^-$ -decay Th- $n$ Tc <sup>99m</sup> I.T. U- $n$
Tc <sup>99</sup>	A m.s.		$\beta^-$	$9.4 \times 10^5$ yr; $4.7 \times 10^4$ yr; $\sim 3 \times 10^5$ yr yield	0.32 abs. Al; $\sim 0.4$ abs. Al; $\sim 0.3$ abs. Al	No $\gamma$	Tc <sup>99</sup> - $n-\gamma$ Mo <sup>100</sup> - $d-2n$ Mo- $p-n$ Mo- $p-n$ Mo <sup>101</sup> $\beta^-$ -decay U- $n$ , Mo <sup>101</sup> $\beta^-$ -decay Ru- $\gamma-p$ U- $n$ , Mo <sup>102</sup> $\beta^-$ -decay Ru- $n-p$ U- $n$ , Mo <sup>106</sup> $\beta^-$ -decay, parent of Ru <sup>106</sup>
Tc <sup>100</sup>	B		$\beta^-$ , $\gamma$	80 sec	2.3 abs. Al	0.6 abs. Pb	Tc <sup>99</sup> - $n-\gamma$ Mo <sup>100</sup> - $d-2n$ Mo- $p-n$ Mo- $p-n$ Mo <sup>101</sup> $\beta^-$ -decay U- $n$ , Mo <sup>101</sup> $\beta^-$ -decay Ru- $\gamma-p$ U- $n$ , Mo <sup>102</sup> $\beta^-$ -decay Ru- $n-p$ U- $n$ , Mo <sup>106</sup> $\beta^-$ -decay, parent of Ru <sup>106</sup>
Tc <sup>&lt;101</sup>	F		$\beta^-$	36.5 hr			
Tc <sup>&lt;101</sup>	E		$\beta^-$	18 sec			
Tc <sup>101</sup>	A		$\beta^-$ , $\gamma$	14.0 min	1.3; 1.1 cl.ch. (K.U.)	0.30	
Tc <sup>102</sup>	D		$\beta^-$	< 1 min			
Tc <sup>&lt;104</sup>	F		K(?), $\gamma$	60 days			
Tc <sup>105</sup>	B		$\beta^-$	Short			
44 Ru <sup>95</sup>	F			20 min			
Ru <sup>95</sup>	A		$\beta^+$ , K, $\gamma$	1.65 hr	1.1( $\beta^+$ ) abs. Al	0.95 abs. Pb	Ru- $n-2n$ (?) Mo- $\alpha-n$ Mo <sup>92</sup> - $\alpha-n$ Ru- $n-2n$ , parent of Tc <sup>95</sup>

Isotope Z      A	Class	Per cent abundance	Type of radiation	Half-life	Energy of radiation, Mev		Produced by
					Particles	$\gamma$ -rays	
44 Ru <sup>96</sup> Ru <sup>97</sup>	A	5.68	K, $\gamma$ , $e^-$	2.8 days; 3.0 days		0.23 abs. Pb	Mo <sup>94</sup> $\alpha$ - $n$ Ru- $d$ - $p$ Ru- $n$ - $\gamma$ , parent of Tc <sup>97m</sup>
Ru <sup>98</sup> Ru <sup>99</sup> Ru <sup>100</sup> Ru <sup>101</sup> Ru <sup>102</sup> Ru <sup>102</sup>	A	2.22 12.81 12.70 16.98 31.34	$\beta^-$ , $\gamma$	42 days; 41 days; 45 days; 37 days	0.25; 0.3 (95 %), 0.8 (5 %) abs. Al; 0.75 abs. Al	0.56 abs. Pb; 0.4 abs. Pb	Ru- $d$ - $p$ Ru- $n$ - $\gamma$ U- $n$ , parent of Rh <sup>103m</sup> U <sup>232</sup> - $n$ Th- $n$ Pu- $n$ Bi- $d$ Pb- $\alpha$
Ru <sup>104</sup> Ru <sup>106</sup>	B	18.27	$\beta^-$ , $\gamma$	4.5 hr; 4.4 hr; 4 hr	1.4 abs. Al; 1.5 abs.; 1.3 abs. Al	0.76 abs. Pb; 0.7 abs. Pb	Ru- $n$ - $\gamma$ Ru- $d$ - $p$ U- $n$ , Tc <sup>105</sup> $\beta^-$ -decay parent of Rh <sup>105</sup> Th- $n$ Bi- $\alpha$ Pb- $\alpha$ Tl- $\alpha$ Pt- $\alpha$
Ru <sup>106</sup>	A m.s.		$\beta^-$	1.0 yr; 290 days	$\sim$ 0.03 abs. Al; very soft	No $\gamma$	U- $n$ , parent of Rh <sup>106</sup> U <sup>232</sup> - $n$ U- $d$ Th- $n$ Th- $\alpha$ Pu- $n$ Bi- $d$
Ru <sup>107</sup>	D		$\beta^-$	4 min	$\sim$ 4 abs. Al		U- $n$ , parent of Rh <sup>107</sup>
45 Rh <sup>100</sup>	B		K, $\gamma$ , $e^-$ , $\beta^+$ ( $\sim$ 5 %)	19.4 hr; 21 hr	0.6( $e^-$ ), 3.0( $\beta^+$ ) spect.	1.2 abs. Pb; 1.8 abs. Pb	Ru- $d$ - $n$ Pd <sup>100</sup> K-decay
Rh <sup>101</sup>	B		K, $\gamma$ , $e^-$	4.3 days; 5.9 days		0.35 abs. Pb, spect. conv.	Ru- $d$ - $n$ Pd <sup>101</sup> K- and $\beta^+$ - decay
Rh <sup>102</sup>	A		$\beta^-$ , $\beta^+$ , $\gamma$ , K	210 days; 215 days	1.04( $\beta^-$ ), 1.13( $\beta^+$ ) cl.ch.; 1.3 abs. Al; 1.1( $\beta^-$ ) abs.	0.46 (annih.?) abs. Pb	Ru- $d$ - $n$ Rh- $n$ - $2n$

Isotope Z A	Class	Per cent abundance	Type of radiation	Half-life	Energy of radiation, Mev		Produced by
					Particles	$\gamma$ -rays	
45 Rh <sup>101</sup> Rh <sup>102m</sup>	A	100	I.T., $e^-$	57 min; 52 min; 48 min; 45 min	0.034( $e^-$ ) spect.; $\sim 0.03(e^-)$ abs. Al	0.040 abs. argon of $e^-$ ; 0.042 abs. of $e^-$	Rh- $n$ - $n$ Rh- $e^-$ - $e^-$ Rh-x-rays Pd <sup>101</sup> K-decay U- $n$ , Ru <sup>102</sup> $\beta^-$ -decay
Rh <sup>104m</sup>	A		I.T., $\gamma$ , $e^-$	4.2 min; 4.4 min; 4.7 min		0.069 spect. conv.; 0.087; 0.09(A38) abs. Al	Ru- $p$ - $n$ Rh- $n$ - $\gamma$ ( $\sim 10\%$ ) Pd- $\gamma$ - $p$
Rh <sup>104</sup>	A		$\beta^-$ , $\gamma$ , $e^-$	44 sec	2.3 cl.ch.; 2.6 spect.; 2.3 abs. Al	0.041, 0.18, 0.95 abs., abs. of $e^-$	Ru- $p$ - $n$ Rh- $n$ - $\gamma$ ( $\sim 90\%$ ) Rh <sup>104m</sup> I.T.
Rh <sup>105</sup>	A		$\beta^-$ , $\gamma$ , $e^-$	36.5 hr; 37 hr; 34 hr	0.65 abs. Al; 0.78 abs. Al; 0.5 abs.	0.33(weak) abs. Pb	Ru- $d$ - $n$ Ru <sup>105</sup> $\beta^-$ -decay Rh- $t$ - $p$ Pd- $\gamma$ - $p$ U- $n$ , Ru <sup>105</sup> $\beta^-$ -decay Th- $n$ Pu- $n$
Rh <sup>106</sup>	A		$\beta^-$ , $\gamma$	30 sec	3.55(82 %), 2.30 (18 %) spect., coincid. abs.; 3.9 (80 %), 2.8(20 %) abs. Al, coincid. abs.; 4.5 abs. Al $\sim 1.3$ abs. Al	1.25(1 %), 0.73 (17 %), 0.51 (17 %) spect.; 0.3 (20 %), 0.8(20 %) abs. Pb	U- $n$ , Ru <sup>106</sup> $\beta^-$ -decay Pu- $n$
Rh	E		$\beta^-$ , $\gamma$	9 hr	$\sim 1.3$ abs. Al	0.8 abs. Pb	U- $n$
Rh <sup>107</sup>	D		$\beta^-$	24 min	1.2 abs. Al		U- $n$ , Ru <sup>107</sup> $\beta^-$ -decay
46 Pd <sup>100</sup>	B		K, $\gamma$	4.0 days		0.090, 1.8 abs. Al, Ag, Pb	Rh- $d$ -5 $n$ Sb- $d$ -5 $\alpha$ 23 $\alpha$ , parent of Rh <sup>100</sup>
Pd <sup>101</sup>	B		K( $\sim 90\%$ ); $\beta^+$ ( $\sim 10\%$ )	9 hr	2.3( $\beta^+$ ) spect.	No $\gamma$	Rh- $d$ -4 $n$ Sb- $d$ -6 $\alpha$ 22 $\alpha$ , parent of Rh <sup>101</sup>
Pd <sup>102</sup> Pd <sup>103</sup>	A	0.8	K	17 days			Rh- $d$ -2 $n$ Rh- $p$ - $n$

Isotope Z     A	Class	Per cent abundance	Type of radiation	Half-life	Energy of radiation, Mev		Produced by
					Particles	$\gamma$ -rays	
46 Pd <sup>103</sup>							Pd- $n$ - $\gamma$ , parent of Rh <sup>103m</sup>
Pd <sup>104</sup>		9.3					
Pd <sup>105</sup>		22.6					
Pd <sup>106</sup>		27.2					
Pd <sup>108</sup>		26.8					
Pd <sup>109</sup>	A m.s.		$\beta^-$	13 hr	1.03 cl.ch.; 1.0 abs. Al; 1.1 abs. Al	No $\gamma$	Pd- $\gamma$ - $n$ Pd- $d$ - $p$ Pd- $n$ - $\gamma$ Ag- $n$ - $p$ Ag- $d$ -2 $p$ Ag- $t$ -He <sup>3</sup> U- $n$ , parent of Ag <sup>109m</sup> U <sup>232</sup> - $n$ Pu- $n$
Pd <sup>110</sup>							
Pd <sup>111</sup>	A	13.5	$\beta^-$	26 min	3.5 abs.		Pd- $d$ - $p$ Pd- $n$ - $\gamma$ U- $n$ , parent of Ag <sup>111</sup> Th- $n$
Pd <sup>112</sup>	A		$\beta^-$	21 hr	0.2 abs. Al	No $\gamma$	U- $n$ , parent of Ag <sup>112</sup> Th- $n$ Th- $\alpha$ Bi- $d$ Pu- $n$ Pd- $p$ - $n$ Sb- $d$ -21 $\alpha$ 5z Pd- $p$ - $n$ Pd- $p$ - $n$
47 Ag <sup>102,104</sup>	C		$\beta^+$ ; K	73 min			
Ag <sup>104</sup>	E			16.3 min			
Ag <sup>105</sup>	E		K, $\gamma$	45 days		0.282, 0.345, 0.430, 0.650, > 1.0 spect.; 0.29, 0.42, 0.50, 0.62 spect.	
Ag <sup>106</sup>	A		$\beta^+$	24.5 min	2.04 abs.	No $\gamma$	Rh- $\alpha$ - $n$ Pd- $d$ - $n$ Pd- $p$ - $\gamma$ Pd- $p$ - $n$ Ag- $n$ -2 $n$ Ag- $d$ - $t$ Ag- $\gamma$ - $n$ Ag- $e^-$ - $e^-$ - $n$ Ag- $d$ - $p$ 2 $n$ Cd- $n$ - $p$ Rh- $\alpha$ - $n$ Pd- $d$ - $n$ Pd- $p$ - $n$
Ag <sup>106</sup>	A		K, $e^-$ , $\gamma$	8.2 days	1.2( $e^-$ ) abs.	1.06, 0.69 spect.; 1.63, 1.06,	

Isotope Z      A	Class	Per cent abundance	Type of radiation	Half-life	Energy of radiation, Mev		Produced by
					Particles	$\gamma$ -rays	
47 Ag <sup>106</sup>						0.72 (?) spect.	Ag-n-2n Ag-d-p2n(?) Cd-n-p Sn-d-?
Ag <sup>107</sup> Ag <sup>107m</sup>	A	51.35	I.T., $e^-$ , $\gamma$	44.3 sec; 40 sec		0.093 spect. conv.; 0.094 spect. conv.	Ag-n-n Ag-x-rays Ag-e <sup>-</sup> -e <sup>-</sup> Cd <sup>107</sup> K-decay
Ag <sup>108</sup>	A		$\beta^-$	2.3 min; 2.4 min	2.8 cl.ch.		Pd-p-n Ag-n- $\gamma$ Ag- $\gamma$ -n Ag-e <sup>-</sup> -e <sup>-</sup> -n Ag <sup>107</sup> -n- $\gamma$ Ag-d-p Cd-n-p Pd <sup>108</sup>
Ag <sup>109m</sup>	A		I.T., $e^-$ , $\gamma$	40.4 sec; 40 sec; 39.2 sec		0.087 spect. conv.; 0.088 spect. conv.	$\beta^-$ -decay Ag-n-n Ag-x-rays Ag-e <sup>-</sup> -e <sup>-</sup> Cd <sup>109</sup> K-decay
Ag <sup>109</sup> Ag <sup>110</sup>	A	48.65	$\beta^-$ , $\gamma$	24.2 sec; 22 sec; 28 sec	2.6 abs.; 2.8 cl.ch. (K.U.)		Ag-n- $\gamma$ Ag <sup>109</sup> -n- $\gamma$ Cd-n-p Cd- $\gamma$ -p Ag-n- $\gamma$ Ag <sup>109</sup> -n- $\gamma$ Ag-d-p
Ag <sup>110</sup>	A res.n.act.		K, $\gamma$ , $e^-$ ; $\beta^-$	225 days	1.3 abs. Al; 0.38 abs. Al; 0.59 spect.	1.40(9 %), 0.90 (47 %), 0.66 (44 %) spect conv., spect.; 0.650, 0.925, 1.51 spect.; 0.6 abs. Al No $\gamma$	Pd-d-n Pd- $\alpha$ -p Cd-n-p Cd- $\gamma$ -p U-n, Pd <sup>111</sup> $\beta^-$ -decay U <sup>232</sup> -n U- $\alpha$ Th- $\alpha$ Pu-n Bi-d Cd-n-p Cd- $\gamma$ -p
Ag <sup>111</sup>	A		$\beta^-$	7.5 days	$\sim$ 0.24(?), 1.0 abs., $\sim$ 0.8 abs.		
Ag <sup>112</sup>	A		$\beta^-$ , $\gamma$	3.2 hr	3.6 abs. Al; 2.2 cl.ch.	0.86 abs. Al	



Isotope Z      A	Class	Per cent abundance	Type of radiation	Half-life	Energy of radiation, Mev		Produced by
					Particles	$\gamma$ -rays	
47 Ag <sup>113</sup>							In- $n$ - $\alpha$ U- $n$ , Pd <sup>113</sup> $\beta$ -decay U <sup>232</sup> - $n$ U- $\alpha$ U- $n$ Cd <sup>114</sup> - $\gamma$ - $p$ U- $n$
Ag <sup>113</sup>	A		$\beta^-$	5.3 hr	2.2 abs. Al; 2.0 abs. Al	No $\gamma$	
Ag	E		$\beta^-$ , $\gamma$	22 min	$\sim 3$ abs. Al		
48 Cd <sup>106, 107</sup>	D		$\beta^+$	33 min			Cd- $n$ -2 $n$
Cd <sup>106</sup>		1.215					
Cd <sup>107</sup>	A		K ( $\sim 100\%$ ), $\gamma$ (4%), $\beta^+$ (0.3%)	6.7 hr	0.32 ( $\beta^+$ ) spect.	0.84 (weak) spect.; 0.53 abs. Pb; 0.7 abs.	Ag- $p$ - $n$ Ag- $d$ -2 $n$ Ag- $\alpha$ - $p$ 3 $n$ Cd <sup>106</sup> - $n$ - $\gamma$ Sb- $d$ -16 $\alpha$ 4s or Sb- $d$ -18 $\alpha$ 4s Sn- $d$ -?
Cd <sup>108</sup>		0.875					
Cd <sup>109</sup>	A		K	330 days			Ag- $d$ -2 $n$ Ag- $\alpha$ - $p$ 3 $n$ Cd <sup>108</sup> - $n$ - $\gamma$ Sn- $d$ -? Sb- $d$ -14 $\alpha$ 4s or Sb- $d$ -16 $\alpha$ 4s
Cd <sup>110</sup>		12.39					
Cd <sup>111m</sup>	A		I.T., $e^-$	48.7 min		0.148, 0.247 spect. conv.; 0.195 abs. of $e^-$ ; 0.145, 0.230 spect. conv., spect.	Pd- $\alpha$ - $n$ Ag- $\alpha$ - $p$ 3 $n$ Cd- $n$ - $n$ or Cd- $n$ - $\gamma$ Cd- $x$ -rays Cd- $e^-$ - $e^-$ Cd <sup>110</sup> - $n$ - $\gamma$ U- $n$
Cd <sup>111</sup>		12.75					
Cd <sup>112</sup>		24.07					
Cd <sup>112m</sup>	A	12.26	I.T.	2.3 min			Cd <sup>112</sup> - $n$ - $n$
Cd <sup>114</sup>		28.86					
Cd <sup>115</sup>	A		$\beta^-$ , $\gamma$	2.33 days; 2.5 days	0.6, 1.13 spect.; 0.55, 1.25 abs. Al; 1.11 spect.	0.65 spect.; 0.55 cl.ch. recoil	Cd- $d$ - $p$ Cd- $n$ - $\gamma$ Cd- $n$ -2 $n$ In- $n$ - $p$ Sb- $d$ -2 $\alpha$ 2 $n$ U- $n$ , parent of In <sup>115m</sup> U <sup>235</sup> - $n$ Th- $\alpha$

Isotope $Z$ $A$	Class	Per cent abundance	Type of radiation	Half-life	Energy of radiation, Mev		Produced by
					Particles	$\gamma$ -rays	
48 Cd <sup>115m</sup>	A		$\beta^-$ , $\gamma$	43 days; 44 days; 40 days	1.85 abs. Al; 1.7 abs. Al; 1.5 abs. Al	0.5 abs. Pb	Cd- $d-p$ Cd- $n-\gamma$ In- $n-p$ Sn- $n-\alpha$ (?) U- $n$ U <sup>235</sup> - $n$ Pu- $n$ Bi- $d$ Th- $\alpha$
Cd <sup>115</sup> Cd <sup>117</sup>	A	7.58	$\beta^-$	170 min; 2.72 hr	1.3-1.7 spect.		Cd- $d-p$ Cd- $n-\gamma$ U- $n$ , parent of In <sup>117</sup>
49 In <sup>108</sup> In <sup>109</sup>	E B		K(?), $\gamma$ K; $\beta^+$ , $\gamma$	$\sim 5$ hr 6.5 hr; 5.2 hr	2( $\beta^+$ )	0.65 0.5	Ag- $\alpha$ -3n Ag- $\alpha$ -2n
In <sup>110</sup>	A m.s.		$\beta^+$	65 min	1.6 spect.		Ag- $\alpha$ -n Cd- $p-n$ Cd- $d-2n$ Sn(4.5 hr) K-decay
In	D		$\beta^+$	72 min	2.2 abs. Be		Ag- $\alpha$ -2n Cd- $p-n$ Cd- $d-n$ In- $n-3n$ Ag- $\alpha$ -n Cd- $d-n$ Cd- $p-n$ In- $n-2n$ , parent of In <sup>112</sup>
In <sup>111</sup>	A m.s.		K, $\gamma$ , $e^-$	2.7 days		0.17, 0.25 spect. conv.	Ag- $\alpha$ -n Cd- $p-n$ Cd- $d-n$ In- $n-3n$ Ag- $\alpha$ -n Cd- $d-n$ Cd- $p-n$ In- $n-2n$ , parent of In <sup>112</sup>
In <sup>112m</sup>	B		I.T., $\gamma$ , $e^-$	20 min; 23 min		0.16 spect. conv.; 0.12 abs. of $e^-$	Ag- $\alpha$ -n Cd- $p-n$ Cd- $d-n$ In- $n-3n$ Ag- $\alpha$ -n In- $n-2n$ In <sup>112m</sup> I.T.
In <sup>112</sup>	B		$\beta^+$ , $\beta^-$ (?)	9 min	1.5( $\beta^+$ ) abs.; 1.7( $\beta^+$ ) cl.ch.; 0.47( $\beta^-$ ?) abs.		Ag- $\alpha$ -n Cd- $p-n$ Cd- $d-n$ In- $n-3n$ Ag- $\alpha$ -n In- $n-2n$ In <sup>112m</sup> I.T.
In <sup>113m</sup>	A		I.T., $\gamma$ , $e^-$	105 min		0.39 spect. conv.	Cd- $p-n$ Cd- $d-n$ In- $x$ -rays Sn <sup>112</sup> K-decay
In <sup>113</sup> In <sup>114m</sup>	A	4.23	I.T., $e^-$	48 days		0.19 spect. conv.; 0.186 spect. conv.	Cd- $p-n$ Cd- $d-n$ In- $n-\gamma$ In- $d-p$ In- $n-2n$ Sn- $d-\alpha$ (?) Cd- $p-n$ In <sup>114m</sup> I.T.
In <sup>114</sup>	A		$\beta^-$	72 sec	1.98 cl.ch.; 1.98 spect.		In- $n-2n$

Isotope Z      A	Class	Per cent abundance	Type of radiation	Half-life	Energy of radiation, Mev		Produced by
					Particles	$\gamma$ -rays	
49 In <sup>114</sup>							In- $\gamma$ - $n$ In <sup>112</sup> - $n$ - $\gamma$
In <sup>115m</sup>	A		I.T., $e^-$ , $\gamma$	4.50 hr; 4.53 hr; 4.1 hr		0.34 spect. conv.; 0.3 abs. Al of $e^-$	Cd- $d$ - $n$ In- $n$ - $n$ In- $p$ - $p$ In- $\alpha$ - $\alpha$ In-x-rays In- $e^-$ - $e^-$ U- $n$ , Cd <sup>115</sup> (2.5 days) $\beta^-$ -decay
In <sup>115</sup> In <sup>116</sup>	A	95.77	$\beta^-$	13 sec	2.8 cl.ch.	No $\gamma$	Cd- $p$ - $n$ In- $n$ - $\gamma$ , (25 %) In- $d$ - $p$ Cd- $p$ - $n$ In- $n$ - $\gamma$ , (75 %) In- $d$ - $p$ Sn- $\gamma$ - $p$
In <sup>116</sup>	A		$\beta^-$ , $\gamma$	54.31 min; 54 min	0.85 spect., cl.ch.	2.32, 1.31, 1.12, 0.428 spect.; 1.8, 1.4, 1.0, 0.6, 0.4, 0.2 cl.ch. re- coil; 2.08 ( $\sim 60$ %), $\sim 1.8$ ( $\sim 40$ %) Be- $\gamma$ - $n$ reaction	Cd- $d$ - $n$ Sn- $\gamma$ - $p$ U- $n$ , Cd <sup>117</sup> $\beta^-$ -decay Pu- $n$
In <sup>117</sup>	A		$\beta^-$	117 min; 1.90 hr	1.73 spect.; 1.95 abs. Al	No $\gamma$	Cd- $d$ - $n$ Sn- $\gamma$ - $p$ U- $n$ , Cd <sup>117</sup> $\beta^-$ -decay Pu- $n$
50 Sn	D		K	4.5 hr			Sb- $d$ -?, parent of In (70 min)
Sn <sup>112</sup> Sn <sup>113</sup>	A	0.90	K, $e^-$ , $\gamma$	105 days; $\sim 70$ days		0.085 spect. conv.; no $\gamma$	Cd- $\alpha$ - $n$ In- $p$ - $n$ In- $d$ -2 $n$ Sn- $d$ - $p$ Sn- $n$ - $\gamma$ Sb- $d$ -10 $\alpha$ 2 $s$ or Sb- $d$ -12 $\alpha$ 2 $s$ , parent of In <sup>112m</sup>
Sn <sup>114</sup> Sn <sup>116</sup> Sn <sup>118</sup> Sn <sup>117</sup> Sn <sup>118</sup>		0.61 0.35 14.07 7.54 23.98					

Isotope $Z$ $A$	Class	Per cent abundance	Type of radiation	Half-life	Energy of radiation, Mev		Produced by
					Particles	$\gamma$ -rays	
50 Sn <sup>&lt;118</sup>	E		$\beta^-$	25 min			Sn- $n$ - $\gamma$ Cd- $\alpha$ - $n$ Cd- $\alpha$ - $n$ Cd- $\alpha$ - $n$ Sb- $d$ - $\alpha$
Sn <sup>&lt;118</sup>	E		$\beta^-$	3 hr			
Sn <sup>119m</sup>	D		I.T., $\gamma$ , $e^-$	13 days; 14 days	0.13( $e^-$ ) spect.	0.17 abs. Pb	
Sn <sup>119</sup>		8.62					
Sn <sup>120</sup>		33.03					
Sn <sup>121</sup>	A		$\beta^-$	28 hr; 26 hr	0.4 abs. Al	No $\gamma$	Sn- $d$ - $p$ Sn- $n$ - $\gamma$ Sn <sup>120</sup> - $d$ - $p$ Th- $\alpha$ U- $n$ U <sup>232</sup> - $n$ Th- $\alpha$
Sn <sup>121, 122</sup>	C		$\beta^-$	130; days; 136 days	1.5-1.6 abs. Al; 1.2	No $\gamma$	U- $n$ U <sup>232</sup> - $n$ Th- $\alpha$
Sn <sup>122</sup>		4.78					
Sn <sup>&gt;120</sup>	D		$\beta^-$	$\sim$ 80 hr; 60 hr	0.76 abs. Al		U- $n$ U- $\alpha$
Sn <sup>123</sup>	D		$\beta^-$ , $\gamma$ (?)	10 days; 11 days; 9 days	2.6 abs. Al; 2.5 abs. Al		Sn- $d$ - $p$ Sn- $n$ - $\gamma$ U- $n$ U <sup>232</sup> - $n$
Sn <sup>124</sup>		6.11					
Sn <sup>126</sup>	B		$\beta^-$ , $\gamma$	10 min; 9 min	$\sim$ 2.2 abs. Al	$\sim$ 0.74 abs. Pb	Sn- $d$ - $p$ Sn- $n$ - $\gamma$ Sn- $d$ - $p$ Sn <sup>124</sup> - $d$ - $t$ Sn- $n$ -2 $n$ Sn <sup>120</sup> - $d$ - $p$ Sn- $d$ - $p$ Sn- $n$ - $\gamma$ (?) U- $n$ U <sup>232</sup> - $n$
Sn <sup>123</sup>	D		$\beta^-$	40 min	$\sim$ 3 abs. Al		U- $n$ U <sup>232</sup> - $n$
Sn <sup>121</sup>	B		$\beta^-$	36 min	1.5 abs. Al		U- $n$ U <sup>232</sup> - $n$
Sn <sup>&lt;126</sup>	D		$\beta^-$	$\sim$ 400 days			U- $n$ , parent of Sb <sup>126</sup> U- $n$
Sn <sup>&gt;120</sup>	E		$\beta^-$	17.5 days	1.7		
Sn <sup>&gt;120</sup>	E		$\beta^-$	7.0 days	1.8		
Sn <sup>126</sup>	D		$\beta^-$ , $\gamma$	70 min; 80 min	0.7 or 2.8 abs. Al	1.2 abs. Pb	U- $n$ , parent of Sb <sup>126</sup> U- $n$
Sn <sup>&gt;126</sup>	D		$\beta^-$	$\sim$ 20 min			
51 Sb <sup>117</sup>	D		K, $e^-$	2.8 hr; 3 hr	0.46( $e^-$ ) abs. Al		Sn- $d$ - $n$ Sn- $p$ - $n$ In- $\alpha$ - $n$ Sn- $d$ - $n$ In- $\alpha$ - $n$ Sn- $p$ - $n$ Te <sup>118</sup> K-decay
Sb <sup>118</sup>	D		K, $\gamma$ , $e^-$	5.1 hr	0.20( $e^-$ ) abs. Al	1.5 abs. Pb	Sn- $d$ - $n$ Sn- $p$ - $n$ Sb- $d$ - $p$ 3 $n$ Te <sup>118</sup> K-decay
Sb <sup>118</sup>	B		$\beta^+$	3.3 min; 3.6 min	3.1 abs. Be		Sn- $d$ - $n$ Sn- $p$ - $n$ Sb- $d$ - $p$ 3 $n$ Te <sup>118</sup> K-decay
Sb <sup>119</sup>	B		K	39 hr		No $\gamma$ , no $e^-$	Sn- $d$ - $n$ Sn- $p$ - $n$ Sb- $d$ - $p$ 3 $n$ Te <sup>118</sup> K-decay
Sb <sup>120</sup>	A		$\beta^+$	17 min	1.53 cl.ch.		Sn- $d$ - $n$ Sn- $p$ - $n$ Sn <sup>120</sup> - $d$ -2 $n$ Sb- $n$ -2 $n$

Isotope Z     A	Class	Per cent abundance	Type of radiation	Half-life	Energy of radiation, Mev		Produced by
					Particles	$\gamma$ -rays	
51 Sb <sup>120</sup>							Sb- $\gamma$ - $n$ Sb- $d$ - $t$ Sb- $p$ - $p$ - $n$ Sn <sup>120</sup> - $d$ -2 $n$ Sb- $d$ - $p$ 2 $n$
Sb <sup>120</sup>	B	57.25	K, $\gamma$ , $e^-$	6.0 days		1.1 abs. Pb	
Sb <sup>121</sup>							
Sb <sup>122m</sup>	A		I.T., $e^-$	3.5 min		0.14 abs. of $e^-$	Sb- $n$ - $\gamma$ Sb <sup>121</sup> - $n$ - $\gamma$
Sb <sup>122</sup>	A	42.75	$\beta^-$ , $\gamma$ , $e^-$	2.8 days	1.36, 1.94 spect; 0.81, 1.64 cl.ch., abs.; 1.19, 1.77 co- incid. abs., abs. Al	0.57 spect. conv.; 0.96 coincid. abs.; 0.80 spect.	Sn- $d$ -2 $n$ Sn- $p$ - $n$ Sb- $d$ - $p$ Sb- $n$ - $\gamma$ Bi- $d$
Sb <sup>123</sup>							
Sb <sup>124</sup>	A		$\beta^-$ , $\gamma$	60 days	2.37, 1.62, 1.00, 0.65, 0.48 spect; spect; spect; coincid. abs., 0.74, 2.45 spect; 2.25, 0.53 spect; 1.53 abs.; 0.654 spect; 0.67, 2.45 coincid. abs.	2.04(weak), 1.708, 0.732, 0.654, 0.608, 0.121 spect., spect. conv.; spect.; 1.72 spect.; 1.82 coincid. abs.; 1.67, 1.71 Be- $\gamma$ - $n$ re- action; 1.70 cl.ch. pair	Sb- $d$ - $p$ Sb- $n$ - $\gamma$ I- $n$ - $\alpha$ Sn- $d$ -2 $n$
Sb <sup>124m</sup>	A		I.T., $\beta^-$ , $\gamma$	21 min		0.02(I.T.) abs. of $e^-$	Sb- $n$ - $\gamma$ Sb <sup>123</sup> - $n$ - $\gamma$
Sb <sup>124m</sup>	A		$\beta^-$ , $\gamma$ ; I.T.	1.3 min	3.2 abs. Al	0.014(I.T.) abs. of $e^-$	Sb- $n$ - $\gamma$ Sb <sup>123</sup> - $n$ - $\gamma$
Sb <sup>125</sup>	A		$\beta^-$ , $\gamma$	2.7 yr; sev- eral yr	0.3(65 %), 0.7(35 %) abs. Al; 0.56	0.55 abs. Pb; 0.6 abs. Pb	Sn- $n$ - $\gamma$ , $\beta^-$ -decay, parent of Te <sup>125m</sup> Sn- $d$ - $n$ U- $n$ U <sup>232</sup> - $n$ Th- $\alpha$ U- $n$ U- $n$ , Sn <sup>126</sup> $\beta^-$ -decay U- $n$ , parent of Te <sup>127</sup> U <sup>232</sup> - $n$ Pu- $n$
Sb <sup>&gt;125</sup>	E		$\beta^-$	28 days	1.86		
Sb <sup>126</sup>	D		$\beta^-$	60 min	2.8 or 0.7 abs. Al		
Sb <sup>127</sup>	A		$\beta^-$ , $\gamma$	93 hr; 90 hr	1.2 abs. Al; 0.8	0.72 abs. Pb	

Isotope Z      A	Class	Per cent abundance	Type of radiation	Half-life	Energy of radiation, Mev		Produced by
					Particles	$\gamma$ -rays	
51 Sb <sup>120</sup>	A		$\beta^-$	4.2 hr			U-n, parent of Te <sup>120</sup>
Sb <sup>122</sup>	B		$\beta^-$	5 min			Pu-n
Sb <sup>123</sup>	A		$\beta^-$	< 10 min			U-n, parent of Te <sup>123</sup>
Sb <sup>124</sup>	B		$\beta^-$	< 10 min			U-n, parent of Te <sup>123</sup>
							Th-n
							U-n, parent of Te <sup>124</sup>
52 Te <sup>&lt;118</sup>	D		$\beta^+$	2.5 hr			Sb-d-?
Te <sup>118</sup>	B		K	6.0 days		No $\gamma$ (?)	Sb-d-5n, parent of Sb <sup>118</sup> (3.3 min)
Te <sup>119</sup>	B		K, $\gamma$ , $e^-$	4.5 days	0.2, 0.5( $e^-$ ) spect.	1.4 abs. Pb	Sb-d-4n, parent of Sb <sup>119</sup> Bi-d
Te <sup>120</sup>							
Te <sup>121m</sup>	A	0.091	I.T., $e^-, \gamma$	143 days; 125 days		0.0365(?), 0.082, 0.0885, 0.159, 0.213 spect. conv.; 0.0820, 0.0883, 0.136, 0.1573, 0.2108 spect. conv.; 0.05 spect. conv., abs. Ag; 0.22 abs. Pb	Sn- $\alpha$ -n Sb-d-2n Sb-p-n
Te <sup>121m</sup>	A		I.T., $\gamma$	$5 \times 10^{-8}$ sec		0.23 co- incid. abs.	Te <sup>121m</sup> (143 days) I.T., par- ent of Te <sup>121</sup>
Te <sup>121</sup>	A		K, $\gamma$	17 days		0.61 abs. Pb; 0.615 spect. conv.	Sb-d-2n Sb-p-n Te <sup>121m</sup> (143 days, $5 \times 10^{-8}$ sec) I.T.
Te <sup>122</sup>		2.49					
Te <sup>123</sup>		0.89					
Te <sup>124</sup>		4.63					
Te <sup>126</sup>		7.01					

Isotope $Z$ $A$	Class	Per cent abundance	Type of radiation	Half-life	Energy of radiation, Mev		Produced by
					Particles	$\gamma$ -rays	
52 Te <sup>125m</sup>	A	18.72	I.T., $e^-$	$\sim 60$ days	$\sim 0.12$ ( $e^-$ ) abs. Al	0.086 spect. conv.	Sb <sup>125</sup> $\beta^-$ -decay I <sup>125</sup> K-decay(?)
Te <sup>126</sup> Te <sup>127m</sup>	A		I.T., $e^-$	90 days			Te- $n\gamma$ Te- $d\beta$ I- $n\beta$ U- $n$ , parent of Te <sup>127</sup> U <sup>232</sup> - $n$ Te- $n\gamma$ Te- $d\beta$ Te- $n-2n$ I- $n\beta$ U- $n$ , Te <sup>127m</sup> I.T. U- $n$ , Sb <sup>127</sup> $\beta^-$ -decay
Te <sup>127</sup>	A		$\beta^-$	9.3 hr	0.76 abs. Al	No $\gamma$	
Te <sup>128</sup> Te <sup>129m</sup>	A	31.72	I.T., $e^-$	32 days		0.102 spect. conv.; no hard $\gamma$	Te- $n\gamma$ Te- $d\beta$ Te- $n-2n$ U- $n$ , parent of Te <sup>129</sup> U <sup>232</sup> - $n$ Te- $n\gamma$ Te- $d\beta$ Te- $\gamma-n$ Te- $n-2n$ U- $n$ , Te <sup>129m</sup> I.T. U- $n$ , Sb <sup>129</sup> $\beta^-$ -decay Th- $n$
Te <sup>129</sup>	A	34.46	$\beta^-$ , $\gamma$	72 min	1.8 spect.	0.3, 0.8 abs. Pb	
Te <sup>130</sup> Te <sup>131m</sup>	A		I.T., $e^-$	30 hr		0.177 spect. conv.	Te- $n\gamma$ Te- $d\beta$ U- $n$ , parent of Te <sup>131</sup> Te- $d\beta$ Te- $n\gamma$ U- $n$ , Te <sup>131m</sup> I.T., par- ent of I <sup>131</sup>
Te <sup>131</sup>	A		$\beta^-$	25 min			U- $n$ , Sb <sup>132</sup> $\beta^-$ -decay, parent of I <sup>132</sup> Th- $n$ Th- $\alpha$ Pu- $n$ U- $n$ , parent of I <sup>132</sup> Pu- $n$
Te <sup>132</sup>	B		$\beta^-$ , $\gamma$	77 hr	0.36 abs. Al; $\sim 0.3$ abs.	0.22 abs. Pb	
Te <sup>133</sup>	A		$\beta^-$	60 min			

Isotope Z     A	Class	Per cent abundance	Type of radiation	Half-life	Energy of radiation, Mev		Produced by
					Particles	$\gamma$ -rays	
52 Te <sup>124</sup>	B		$\beta^-$	43 min			U-n, Sb <sup>124</sup> $\beta^-$ -decay, parent of I <sup>124</sup>
Te <sup>125</sup>	A		$\beta^-$	< 2 min			Th-n
Te	D		$\beta^-$	~ 1 min			Pu-n U-n, parent of I <sup>125</sup>
53 I <sup>124</sup>	A		$\beta^+$	4.0 days			U-n
I <sup>125</sup>	B		K, no $\beta$	56 days	~ 0.1 (weak) ( $e^-$ ?)	No $\gamma$ , no $e^-$	Sb- $\alpha$ -n Te- $p$ -n Bi-d
I <sup>126</sup>	A		$\beta^-, \gamma$	13.0 days	1.1 abs.	0.5 abs. Pb	Te-d-n Bi-d Sb- $\alpha$ -n Te-d-n Te- $p$ -n I-n-2n I- $\gamma$ -n Bi-d
I <sup>127</sup> I <sup>128</sup>	A	100	$\beta^-, \gamma$	24.99 min	1.59 (7 %) (by diff.), 2.02 (93 %) spect.; 1.05, 2.10 cl.ch. (K.U.)	0.428 (7 %) spect.; 0.4 abs. Pb	I-n- $\gamma$ Te-d-2n Te- $p$ -n
I <sup>129</sup> I <sup>130</sup>	A A		$\beta^-$ $\beta^-, \gamma$	Long 12.6 hr	0.61, 1.03 spect. coincid.	0.417, 0.537, 0.667, 0.744 spect. conv., spect., coincid.	U-n Te-d-2n Te- $p$ -n Cs-n- $\alpha$ Th-n(?) I <sup>129</sup> -n- $\gamma$
I <sup>131</sup>	A		$\beta^-, \gamma, e^-$	8.0 days	0.595 spect., coincid.; 0.687 cl.ch.	0.367, 0.080 spect., spect. conv., coincid.; 0.65 (15 %) abs.; 0.4 abs. Pb	Te-d-n U-n, Te <sup>131</sup> $\beta^-$ -decay U <sup>232</sup> -n U- $\alpha$ Th- $\alpha$ Pu-n
I <sup>132</sup>	B		$\beta^-, \gamma$	2.4 hr	0.9, 2.2 abs. Al; ~ 1.35 abs.	0.6, 1.4 abs. Pb; 0.85 abs.	U-n, Te <sup>132</sup> $\beta^-$ -decay, parent of Xe <sup>132</sup> U <sup>232</sup> -n U- $\alpha$ Th-n



Isotope Z     A	Class	Per cent abundance	Type of radiation	Half-life	Energy of radiation, Mev		Produced by
					Particles	$\gamma$ -rays	
53 I <sup>123</sup>	A		$\beta^-$ , $\gamma$	22 hr; 20.5 hr	1.4 abs. Al; 1.1 cl.ch.	0.55 abs. Pb; 0.528 spect.	U- $\pi$ , Te <sup>123</sup> $\beta^-$ -decay, parent of Xe <sup>123</sup> U- $\alpha$ Pu- $\pi$ Pb- $\alpha$
I <sup>124</sup>	B		$\beta^-$ , $\gamma$	54 min		> 1 abs. Pb	U- $\pi$ , Te <sup>124</sup> $\beta^-$ -decay Th- $\pi$ U- $\alpha$ Pu- $\pi$
I <sup>125</sup>	A		$\beta^-$ , $\gamma$	6.7 hr; 6.6 hr	1.40(25 %), 1.00 (40 %), 0.47 (35 %) spect.; 1.4 abs. Al; 1.6 abs.	1.6 abs. Pb; 1.3 abs.; 1.27, 2.00 spect.	U- $\pi$ , Te <sup>125</sup> $\beta^-$ -decay, parent of Xe <sup>125</sup> , or parent of Xe <sup>126m</sup> (~ 10 %), Xe <sup>125</sup> (~ 90 %) Th- $\pi$ Pu- $\pi$ U- $\alpha$
I <sup>126</sup>	D		$\beta^-$ , $\gamma$	1.8 min; 86 sec	6.5 abs. Al	2.9 abs. Pb	U- $\pi$ , parent of Xe <sup>126</sup>
I <sup>127</sup>	D		$\beta^-$ , $\pi$	22.0 sec; 22.5 sec; 18 sec	0.56(mean) ( $\pi$ ) abs. paraffin; 0.7(mean) ( $\pi$ ) $\beta$ re- coil in cl.ch.		U- $\pi$ , parent of Xe <sup>127</sup> Pu- $\pi$
I <sup>128</sup>	D		$\beta^-$	5.9 sec			U- $\pi$ , ances- tor of Cs <sup>128</sup>
I <sup>129</sup>	D		$\beta^-$	2.6 sec			U- $\pi$ , ances- tor of Ba <sup>129</sup>
I	F			30 days			Xe- $n$ - $p$
54 Xe <sup>124</sup>		0.094					
Xe <sup>126</sup>		0.088					
Xe <sup>127</sup>	B		I.T.(?), $e^-$ , $\gamma$	75 sec		0.175, 0.125 spect. conv.	I- $p$ - $\pi$
Xe <sup>127</sup>	B		$e^-$ , $\gamma$	34 days		0.9 abs. of $e^-$	Xe- $n$ - $\gamma$ I- $p$ - $\pi$ I- $d$ -2 $\pi$
Xe <sup>128</sup>		1.90					
Xe <sup>129</sup>		26.23					
Xe <sup>130</sup>		4.07					
Xe <sup>131</sup>		21.17					U- $n$ m.s.
Xe <sup>132</sup>		26.96					U- $n$ m.s.
Xe <sup>m</sup>	F		I.T., $e^-$	11 days			Xe- $n$ - $n$

Isotope $Z \quad A$	Class	Per cent abundance	Type of radiation	Half-life	Energy of radiation, Mev		Produced by
					Particles	$\gamma$ -rays	
54 Xe <sup>131</sup>	A		$\beta^-$ , $\gamma$ , $e^-$	5.3 days; 5.4 days	0.34 abs.; 0.049( $e^-$ ) abs.; 0.260 abs. Al; 0.42 abs. Al.	0.085 abs. Cu, Pb	Te- $\alpha$ -n Xe-d-p Xe-n- $\gamma$ Cs-n-p Ba-n- $\alpha$ U-n, I <sup>133</sup> $\beta^-$ -decay U-n m.s.
Xe <sup>134</sup> Xe <sup>135</sup>	A	10.54	$\beta^-$ , $\gamma$ , $e^-$ (10 %)	9.2 hr; 9.4 hr	0.93 spect.; 0.95 abs. Al; 0.9 abs. Al; 1.0 abs. Al	0.247 spect.; 0.25 abs. Pb	Xe-d-p Ba-n- $\alpha$ U-n, I <sup>136</sup> $\beta^-$ -decay, parent of Xe <sup>135m</sup> I.T.
Xe <sup>135m</sup>	A		$\gamma$ ; I.T., $\gamma$ , $e^-$	15.6 min; 10 min		0.52 spect.; ~ 0.5 abs. Pb; 0.6 abs. Al of $e^-$	Xe-n- $\gamma$ U-n, I <sup>136</sup> $\beta^-$ -decay, parent of Xe <sup>136</sup>
Xe <sup>136</sup> Xe <sup>137</sup> Xe <sup>137</sup>	D B	8.95	$\beta^-$	68 min 3.8 min; 3.4 min	4 abs. Al		U-n m.s. Xe-d-p Xe-n- $\gamma$ U-n, I <sup>137</sup> $\beta^-$ -decay, parent of Cs <sup>137</sup>
Xe <sup>138</sup>	D		$\beta^-$	17 min			U-n, parent of Cs <sup>138</sup>
Xe <sup>139</sup>	A		$\beta^-$	41 sec; ~ 0.5 min			U-n, parent of Cs <sup>139</sup>
Xe <sup>140</sup>	A		$\beta^-$	16 sec; < 0.5 min; 9.8 sec			Th-n U-n, ances- tor of Ba <sup>140</sup>
Xe <sup>141</sup>	A		$\beta^-$	1.7 sec			Th-n U-d U-n, ances- tor of Ce <sup>141</sup>
Xe <sup>142</sup>	A		$\beta^-$	~ 1.3 sec			U-d U-n, ances- tor of Pr <sup>142</sup>
Xe <sup>144</sup>	A		$\beta^-$	Short			U-n, ances- tor of Ce <sup>144</sup>
Xe <sup>145</sup>	D		$\beta^-$	0.8 sec; short			U-n, ances- tor of Pr <sup>145</sup>
55 Cs <sup>130</sup> Cs <sup>131</sup>	B B		K, $\gamma$ , $e^-$	30 min 10.2 days; 10.0 days		No $\gamma$ ; 0.145 abs. of $e^-$	I- $\alpha$ -n Ba <sup>131</sup>
Cs <sup>132</sup>	B		K, $\gamma$ , $e^-$	7.1 days	0.6( $e^-$ ) abs. Al	0.62 abs. Pb	K-decay Cs-n-2n
Cs <sup>133</sup> Cs <sup>134m</sup>	A	100	$\beta^-$ ; $\gamma$ ; I.T., $e^-$	3.15 hr; 3 hr	2.4 abs. Al; 1 abs.	0.7 abs. Pb; 0.15 (I.T.) spect. conv.; 0.16 (I.T.) abs. of $e^-$	Cs-n- $\gamma$ Cs-d-p

Isotope Z      A	Class	Per cent abundance	Type of radiation	Half-life	Energy of radiation, Mev		Produced by
					Particles	$\gamma$ -rays	
55 Cs <sup>134</sup>	A		$\beta^-$ , $\gamma$ , $e^-$ (2.5 %)	2.3 yr; 1.7 yr	0.09 (25 %), 0.66 (75 %) spect.; 0.65 spect.; 0.75 abs. Al; 0.64 spect.; 0.9 abs.; 0.8 coincid. abs.	0.57 (25 %), 0.60 (100 %), 0.79 (100 %) spect.; 0.58, 0.78, 1.35 (weak) spect., coincid.; 0.61, 0.80 spect.	Cs-n- $\gamma$ Cs-d- $p$ Ba-d- $\alpha$
Cs <sup>136</sup>	A		$\beta^-$ , $\gamma$	13.7 days; 13 days; 10.2 days	$\sim 0.28$ abs. Al; $\sim 0.35$ abs. Al	0.9 abs. Pb; 1.2 abs. Pb	Ba-n- $p$ La-n- $\alpha$ U <sup>232</sup> -n Pu-n Th- $\alpha$
Cs <sup>137</sup>	A m.s.		$\beta^-$	37 yr yield; 33 yr yield	0.550 (single) spect.; 0.57 abs. Al		Xe-n- $\gamma$ , Xe $\beta^-$ -decay, parent of Ba <sup>137m</sup> U-n U <sup>232</sup> -n Pu-n Th- $\alpha$
Cs <sup>138</sup>	D		$\beta^-$ , $\gamma$	33 min	2.6 abs.	1.2 abs. Pb	Ba-n- $p$ U-n, Xe <sup>138</sup> $\beta^-$ -decay Pa-n Th-n
Cs <sup>139</sup>	A		$\beta^-$	9.7 min; 7 min; 10 min			U-n, Xe <sup>139</sup> $\beta^-$ -decay, parent of Ba <sup>139</sup> Th-n
Cs <sup>140</sup>	D		$\beta^-$	65 sec; 40 sec			U-n
Cs <sup>141</sup>	A		$\beta^-$	Short			U-n, Xe <sup>141</sup> $\beta^-$ -decay, ancestor of Ce <sup>141</sup>
Cs <sup>142</sup>	D		$\beta^-$	Short			U-n, parent of Ba <sup>142</sup>
Cs <sup>143</sup>	A		$\beta^-$	Short			U-n, Xe <sup>143</sup> $\beta^-$ -decay, ancestor of Pr <sup>143</sup>
Cs <sup>144</sup>	A		$\beta^-$	Short			U-n, Xe <sup>144</sup> $\beta^-$ -decay, ancestor of Ce <sup>144</sup>
Cs <sup>145</sup>	D		$\beta^-$	Short			U-n, Xe <sup>145</sup> $\beta^-$ -decay, ancestor of Pr <sup>145</sup>

Isotope <i>Z</i> <i>A</i>	Class	Per cent abundance	Type of radiation	Half-life	Energy of radiation, Mev		Produced by
					Particles	$\gamma$ -rays	
56 Ba <sup>130</sup> Ba <sup>131</sup>	B	0.101	K, $\gamma$ ; no $\beta^+$ , $e^-$	12.0 days; 11.7 days		0.22, 0.50, 1.7(weak); 0.26, 0.5, 1.2(weak) abs. Pb, abs. of $e^-$	Ba- $n$ - $\gamma$ , parent of Cs <sup>131</sup>
Ba <sup>132</sup> Ba <sup>133m</sup>	A	0.097	I.T., $e^-$ , $\gamma$ (?)	38.8 hr; 37.8 hr		0.30 spect. conv.; 0.276 spect. conv.	Cs- $p$ - $n$ Cs- $d$ -2 $n$ Ba- $n$ -2 $n$ Ba- $d$ - $p$ Bi- $\alpha$ Bi- $d$ Pb- $\alpha$ Ba- $n$ - $\gamma$ Ba <sup>133m</sup> I.T.
Ba <sup>133</sup>	A		K, $\gamma$ , $e^-$	> 20 yr		0.36 abs. Pb, abs. of $e^-$ ; 0.085, 0.320 abs., abs. of $e^-$ , cl.ch.	
Ba <sup>134</sup> Ba <sup>135m</sup>	D	2.42	I.T., $\gamma$ , $e^-$	28.7 hr	0.28( $e^-$ ) abs. Al	0.34(weak) abs. Pb	Ba- $n$ - $\gamma$ Ba- $d$ - $p$ U- $\alpha$
Ba <sup>135</sup> Ba <sup>136</sup> Ba <sup>137m</sup>	A	6.59 7.81	I.T., $\gamma$ , $e^-$	2.63 min; 2.5 min	0.626( $e^-$ ) spect., coincid.; 0.7( $e^-$ ) abs. Al, coincid.	0.663 spect. conv., spect.; 0.75 abs. Pb	Cs <sup>137</sup> $\beta^-$ -decay Ba- $n$ - $\gamma$
Ba <sup>137</sup> Ba <sup>138</sup> Ba <sup>139</sup>	A	11.32 71.66	$\beta^-$ , $\gamma$	84 min; 85 min; 86 min	2.27 spect.; 2.3 abs.	0.163, 1.05 spect. conv., abs. Pb, coincid.; 0.6 abs. Pb, Cu	Ba- $d$ - $p$ Ba- $n$ - $\gamma$ La- $n$ - $p$ Ce- $n$ - $\alpha$ U- $n$ , Cs <sup>139</sup> $\beta^-$ -decay U- $\gamma$ Th- $n$ Pu- $n$
Ba <sup>140</sup>	A m.s.		$\beta^-$ , $\gamma$ , $e^-$	308 hr; 12.8 days; 12.5 days	1.05 spect.; 0.4(25 %), 1.0(75 %) abs. Al; 1.2 abs.; 1.1 abs.	0.529 spect.; 0.54 spect., spect. conv.; 0.5(25 %) abs. Pb	U- $n$ , Xe <sup>140</sup> (and Cs <sup>140</sup> ) $\beta^-$ -decay, parent of La <sup>140</sup> U <sup>233</sup> - $n$ U- $d$ U- $\alpha$ Th- $n$ Th- $\alpha$ Pu- $n$

Isotope Z A	Class	Per cent abundance	Type of radiation	Half-life	Energy of radiation, Mev		Produced by
					Particles	$\gamma$ -rays	
56 Ba <sup>141</sup>	A		$\beta^-$ , $\gamma$	18 min			U-n, Cs <sup>141</sup> $\beta^-$ -decay, parent of La <sup>141</sup> Th-n U- $\gamma$
Ba <sup>142</sup>	D		$\beta^-$	6 min			U-n, Cs <sup>142</sup> $\beta^-$ -decay, parent of La <sup>142</sup> Th-n U- $\gamma$
Ba <sup>143</sup>	B		$\beta^-$	< 1 min			U-n, parent of La <sup>143</sup> Th-n
Ba <sup>144</sup>	A		$\beta^-$	Short			U-n, de- scendant of Xe <sup>144</sup> , ancestor of Ce <sup>144</sup>
Ba <sup>145</sup>	D		$\beta^-$	Short			U-n, de- scendant of Xe <sup>145</sup> , ancestor of Pr <sup>145</sup>
57 La <sup>&lt;139</sup> La <sup>139</sup>	D B		$\beta^+$ K, $\gamma$	10 min 19.5 hr; 17.5 hr	2.1 abs. Al	0.88 abs. Pb	Ba-d-n Cs- $\alpha$ -2n Ba-d-n Ba-p-n Ce <sup>139</sup> $\beta^+$ -decay
La <sup>136</sup> La <sup>137</sup>	B A		$\beta^+$	2.1 hr > 400 yr	0.84 abs. Al	No $\gamma$	Cs- $\alpha$ -n Ce <sup>137</sup> K-decay
La <sup>138</sup> La <sup>139</sup> La <sup>140</sup>	m.s. A m.s.	0.089 99.911	$\beta^-$ , $\gamma$	40.4 hr; 40.0 hr; 39.5 hr	0.90(20 %), 1.40 (70 %), 2.12 (10 %) spect.; 1.41 abs. Al, spect.; 1.45 spect.; 1.8 abs.	0.335(2 %), 0.49(5 %), 0.87 (10 %), 1.65 (77 %), 2.3(6 %) spect.; 0.335 (1 %), 0.49 (7 %), 0.83 (14 %), 1.63 (74 %), 2.3 (4 %) spect.; 2.49(weak) D- $\gamma$ -n reaction	Ba-d- $\gamma$ (?) La-d-p La-n- $\gamma$ Ce-n-p U-n, Ba <sup>140</sup> $\beta^-$ -decay Th-n Pu-n

Isotope Z      A	Class	Per cent abundance	Type of radiation	Half-life	Energy of radiation, Mev		Produced by
					Particles	$\gamma$ -rays	
57 La <sup>141</sup>	A		$\beta^-$	3.7 hr; 3.5 hr	2.9 abs. Al	No $\gamma$ (?)	U- <i>n</i> , Ba <sup>141</sup> $\beta^-$ -decay, parent of Ce <sup>141</sup> Th- <i>n</i>
La <sup>142</sup>	D		$\beta^-$ , $\gamma$	74 min; 77 min			U- <i>n</i> , Ba <sup>142</sup> $\beta^-$ -decay Th- <i>n</i>
La <sup>143</sup>	A		$\beta^-$	20 min; 15 min			U- <i>n</i> , Ba <sup>143</sup> $\beta^-$ -decay, parent of Ce <sup>143</sup>
La <sup>144</sup>	A		$\beta^-$	Short			U- <i>n</i> , de- scendant of Xe <sup>144</sup> , parent of Ce <sup>144</sup>
La <sup>145</sup>	D		$\beta^-$	Short			U- <i>n</i> , de- scendant of Xe <sup>145</sup> , ancestor of Pr <sup>145</sup>
58 Ce <sup>135</sup>	B		$\beta^+$	~ 16 hr			La- <i>d-6n</i> , parent of La <sup>135</sup>
Ce <sup>136</sup> Ce <sup>137</sup>	B	0.193	K, $\gamma$ , $e^-$	36 hr		0.28, 0.75 abs. Pb	La- <i>d-4n</i>
Ce <sup>138</sup> Ce <sup>139</sup>	B	0.250	K, $\gamma$ , $e^-$	140 days		0.18, 1.8 abs. Pb; 0.18, ~ 0.8 abs. Pb	Ba- $\alpha$ -2 <i>n</i> La- <i>d-2n</i> Bi- <i>d</i>
Ce <sup>140</sup> Ce <sup>141</sup>	A m.s.	88.48	$\beta^-$ , $\gamma$	28 days; 30.6 days	0.60 abs. Al; 0.66 abs. Al; 0.4 abs. Al	0.21 abs. Pb; 0.2	Be- $\alpha$ - <i>n</i> Ce- <i>d-p</i> Ce- <i>n-γ</i> Ce- <i>n-2n</i> Pr- <i>n-p</i> U- <i>n</i> , La <sup>141</sup> $\beta^-$ -decay Th- <i>n</i> Pu- <i>n</i> U- <i>d</i>
Ce <sup>142</sup> Ce <sup>143</sup>	A	11.07	$\beta^-$ , $\gamma$	33 hr; 36 hr	1.36 abs. Al; 1.3 abs. Al	0.5 abs. Pb; 0.6 abs. Pb	Ce- <i>d-p</i> Ce- <i>n-γ</i> U- <i>n</i> , La <sup>143</sup> $\beta^-$ -decay, parent of Pr <sup>143</sup> U- <i>d</i> Th- <i>n</i> Th- $\alpha$ Pu- <i>n</i>

Isotope Z      A	Class	Per cent abundance	Type of radiation	Half-life	Energy of radiation, Mev		Produced by
					Particles	$\gamma$ -rays	
58 Ce <sup>144</sup>	A m.s.		$\beta^-$ , $e^-$	275 days; 300 days	0.348 spect.; 0.25 abs.; 0.30 spect., 0.075, 0.12( $e^-$ ) spect.	No $\gamma$	U- $n$ , de- scendant of Xe <sup>144</sup> , parent of Pr <sup>144</sup> U <sup>232</sup> - $n$ U- $d$ Pu- $n$ Th- $\alpha$ U- $n$ , de- scendant of Xe <sup>148</sup> , parent of Pr <sup>148</sup>
Ce <sup>145</sup>	D		$\beta^-$	1.8 hr			U- $n$ , parent of Pr <sup>146</sup>
Ce <sup>146</sup>	D		$\beta^-$	14.6 min; 11 min			
59 Pr <sup>140</sup>	A		$\beta^+$	3.5 min	2.5 abs. Al; 2.40 cl.ch.		Pr- $n$ -2 $n$ Pr- $\gamma$ - $n$
Pr <sup>141</sup> Pr <sup>142</sup>	A	100	$\beta^-$ , $\gamma$	19.3 hr; 19.2 hr	2.14 spect.; 2.23 spect.	1.9 abs. Pb; $\sim 1.3$ , $\sim 1.65$ spect.	La- $\alpha$ - $n$ Ce- $p$ - $n$ Pr- $d$ - $p$ Pr- $n$ - $\gamma$ Nd- $n$ - $p$ Ce <sup>142</sup> $\beta^-$ -decay U- $n$ , Ce <sup>143</sup> $\beta^-$ -decay U- $d$ Pu- $n$
Pr <sup>143</sup>	A m.s.		$\beta^-$	13.8 days; 13.5 days; 14.2 days; 12.7 days	0.95 abs. Al; 1.0 abs. Al; 0.83 abs. Al	No $\gamma$	U- $n$ , Ce <sup>144</sup> $\beta^-$ -decay U- $d$ Pu- $n$
Pr <sup>144</sup>	A		$\beta^-$ , $\gamma$ , $e^-$	17.5 min; 17 min; 18 min	3.07 spect.; 3.1 abs. 2.99 spect.	0.135 spect. conv.; 1.25, 0.22 abs. Pb	U- $n$ , Ce <sup>145</sup> $\beta^-$ -decay U- $d$ Pu- $n$
Pr <sup>145</sup>	D		$\beta^-$	4.5 hr	3.2 abs. Al	No $\gamma$	U- $n$ , Ce <sup>146</sup> $\beta^-$ -decay U- $n$ , Ce <sup>146</sup> $\beta^-$ -decay
Pr <sup>146</sup>	D		$\beta^-$ , $\gamma$	24.6 min; 25 min	$\sim 3$ abs. Al	1.4 abs. Pb	U- $n$ , Ce <sup>147</sup> $\beta^-$ -decay
60 Nd <sup>141</sup>	B		$\beta^+$ (3 %); K(97 %), $\gamma$	2.42 hr; 2.5 hr	0.78; 0.7 abs. Al	1.05 abs. Pb	Pr- $p$ - $n$ Nd- $d$ - $i$ (?) Nd- $n$ -2 $n$ Nd- $\gamma$ - $n$
Nd <sup>142</sup> Nd <sup>143</sup> Nd <sup>144</sup> Nd <sup>145</sup> Nd <sup>146</sup> Nd <sup>147</sup>	A	27.13 12.20 23.87 8.30 17.18	$\beta^-$ , $\gamma$ , $e^-$	11.0 days; 11.1 days; 12.1 days	0.4(40 %), 0.9(60 %), 0.03( $e^-$ ) abs. Al; 0.76 abs.	0.58(40 %) abs. Pb coincid.; 0.45 abs.	Nd- $n$ - $\gamma$ , parent of Pm <sup>147</sup> U- $n$

Isotope Z      A	Class	Per cent abundance	Type of radiation	Half-life	Energy of radiation, Mev		Produced by
					Particles	$\gamma$ -rays	
60 Nd <sup>148</sup> Nd <sup>149</sup>	B	5.72	$\beta^-$ , $\gamma$ (?)	1.7 hr; 2.0 hr	1.6 abs. Al; 1.5 abs. Al		Nd-n- $\gamma$ Nd-d-p Nd-n-2n, parent of Pm <sup>149</sup> (?)
Nd <sup>150</sup> Nd <sup>150</sup>	E	5.60	$\beta^-$	$\sim 5 \times 10^{10}$ yr	0.011 abs. air		Natural source
Nd <sup>151</sup>	E		$\beta^-$	21 min			Nd-n- $\gamma$
Nd <sup>151</sup>	F		$\beta^-$	Short			Nd-n- $\gamma$ , parent of Pm <sup>151</sup>
61 Pm <sup>143</sup>	B		K, $e^-$ , $\gamma$	$\sim 200$ days; $\sim 1$ yr		0.67 abs.	Pr- $\alpha$ -2n Nd-d-n Nd-p-n Nd-d-n Nd- $\alpha$ -p Nd-d-n
Pm	E		$\beta^-$ , $\gamma$	2.7 hr	2		U-n U <sup>232</sup> -n Nd-n- $\gamma$ , Nd <sup>147</sup> $\beta^-$ - decay
Pm Pm <sup>147</sup>	E A m.s.		$\beta^-$ , $\gamma$ $\beta^-$	16 days 3.7 yr; $\sim 4$ yr; 2-3 yr	1.7 0.223 spect.; $\sim 0.2$ abs. Al; 0.20 abs. Al	No $\gamma$	Pm <sup>147</sup> -n- $\gamma$ Nd-p-n Nd-d-2n Nd- $\alpha$ -p Nd-n- $\gamma$ , Nd $\beta^-$ -decay U-n Pu-n Nd-d-n Nd-n- $\gamma$ , Nd <sup>152</sup> $\beta^-$ - decay
Pm <sup>148</sup>	A m.s.		$\beta^-$ , $\gamma$	5.3 days	2.5 abs.; 2	0.8 abs.	
Pm <sup>149</sup>	A m.s.		$\beta^-$ , $\gamma$	47 hr; 47.5 hr; 55 hr	1.1 abs. Al	0.25(weak) abs. Pb	
Pm Pm <sup>151</sup>	F F		$\beta^-$ $\beta^-$	12.5 hr 12 min			
62 Sm <sup>144</sup> Sm <sup>145</sup>	F m.s.	3.16		$> 150$ days; $> 72$ days		0.242, 0.95 spect. conv., abs. Pb	Sm-n- $\gamma$
Sm <sup>147</sup> Sm <sup>148</sup> Sm <sup>149</sup> Sm <sup>150</sup> Sm <sup>151</sup>		15.07 11.27 13.84 7.47					
Sm <sup>152</sup>	A m.s.		$\beta^-$	$\sim 20$ yr	0.06 abs. Al	No $\gamma$ (?)	Sm-n- $\gamma$ U-n
Sm <sup>152</sup> Sm <sup>153</sup>	B m.s.	26.63	$\alpha$	$1.0 \times 10^{12}$ yr (total Sm); $1.2$ $\times 10^{12}$ yr (total Sm)	2.14 photo- film track; 2.0 cl.ch.		Natural source



Isotope <i>Z</i> <i>A</i>	Class	Per cent abundance	Type of radiation	Half-life	Energy of radiation, Mev		Produced by
					Particles	$\gamma$ -rays	
62 Sm <sup>152</sup>	A m.s.		$\beta^-$ , $\gamma$ ; $e^-$	47 hr; 46 hr	0.78 abs. Al	0.0695, 0.103 spect. conv.; 0.57 (weak), 0.10 abs. Pb, Cu; ~ 0.6, 0.11 spect.; 0.61 (weak), 0.11 abs., coincid. abs.	Nd- $\alpha$ - $n$ Sm- $n$ - $\gamma$ Sm- $n$ -2 $n$ Sm- $d$ - $p$ Sm- $\gamma$ - $n$ U- $n$ U <sup>232</sup> - $n$ Pu- $n$
Sm <sup>154</sup> Sm <sup>156</sup>	B	22.53	$\beta^-$ , $\gamma$	25 min; 21 min	1.9 abs. Al; 1.8	~ 0.3 abs. Pb	Nd- $\alpha$ - $n$ Sm- $n$ - $\gamma$ Sm- $d$ - $p$ U- $n$ U- $n$ , parent of Eu <sup>154</sup>
Sm <sup>158</sup>	A		$\beta^-$	~ 10 hr	~ 0.8 abs. Al		Sm- $d$ - $n$
63 Eu <sup>147</sup>	D			53 days; 40 days			Sm- $d$ - $n$
Eu <sup>149</sup>	D			14 days			Sm- $d$ - $n$
Eu <sup>150</sup>	E		$\beta^+$	27 hr			Eu- $n$ -2 $n$ (?)
Eu <sup>151</sup> Eu <sup>152</sup>	A m.s.	47.77	$\beta^-$ , $\gamma$ , $e^-$ ; K	9.2 hr; 9.3 hr	1.88( $\beta^-$ ) spect.; 0.36, 1.8( $\beta^-$ ) abs. Al	0.123, 0.163, 0.725 spect. conv.; 1.0 abs. Pb	Eu- $n$ - $\gamma$ Eu- $n$ -2 $n$ Eu- $d$ - $p$
Eu <sup>152</sup>	A m.s.		$\beta^-$ , $\gamma$ , $e^-$	Long	0.75( $\beta^-$ ) spect.		Eu- $n$ - $\gamma$
Eu <sup>153</sup> Eu <sup>154</sup>	A m.s.	52.23	$\beta^-$ , $\gamma$ ; K	> 20 yr; 5-8 yr	0.9 spect.; 0.34, 0.84 abs. Al; 0.62, 1.0 coincid. abs.; 1.4 abs. Al; 1.0 abs. Al	1.1 abs. Pb; 0.040, 0.122, 0.247, 0.286, 0.343, 0.408, 1.23 spect. conv., abs.; 0.9 abs. Pb	Sm- $d$ -2 $n$ (?) Eu- $n$ - $\gamma$ Eu- $d$ - $p$
Eu <sup>155</sup>	A m.s.		$\beta^-$ , $\gamma$	2-3 yr	0.18 abs. Al; 0.23 abs. Al	0.084 abs. Al, crit. abs. Tl, Hg	Sm- $n$ - $\gamma$ , Sm <sup>154</sup> $\beta^-$ -decay U- $n$ Th- $\alpha$

Z	Isotope A	Class	Per cent abundance	Type of radiation	Half-life	Energy of radiation, Mev		Produced by
						Particles	$\gamma$ -rays	
63	Eu <sup>156</sup>	A m.s.		$\beta^-$ , $\gamma$	15.4 days	0.5(60 %), 2.5(40 %) abs. Al	2.0(60 %) abs. Pb	Eu <sup>156</sup> , $n$ - $\gamma$ U- $n$ , Sm <sup>156</sup> $\beta^-$ -decay Pu- $n$ Th- $\alpha$ U- $n$ Th- $\alpha$ Pu- $n$
	Eu <sup>157</sup>	D		$\beta^-$ , $\gamma$	15.4 hr	$\sim 1.0$ ( $\sim 75$ %), $\sim 1.8$ ( $\sim 25$ %) abs. Al $\sim 2.5$ abs. Al	0.2, 0.6 abs. Pb	U- $n$ Th- $\alpha$ Pu- $n$
	Eu <sup>&gt;154</sup>	D		$\beta^-$	60 min			U- $n$
64	Gd <sup>152</sup> Gd <sup>153</sup>	B m.s.	0.20	K, $e^-$ , $\gamma$	155 days; 155-170 days; $\sim 110$ days	0.22, 0.40 (weak)( $e^-$ ) abs. Al	0.102 spect. conv.; 0.083, 0.270 abs. Cu, Pb	Eu- $d$ -2 $n$ Gd- $n$ - $\gamma$
	Gd <sup>154</sup> Gd <sup>155</sup> Gd <sup>156</sup> Gd <sup>157</sup> Gd <sup>158</sup> Gd <sup>160</sup> Gd <sup>161</sup>	D	2.15 14.78 20.59 15.71 24.78 21.79	$\beta^-$ , $\gamma$	18.0 hr; 20 hr 8.6 days 4.5 min	0.85	0.3	Gd- $n$ - $\gamma$ Gd- $d$ - $p$ Gd- $n$ - $\gamma$ Gd- $n$ - $\gamma$
	Gd <sup>161</sup>	D		$\beta^-$ , $\gamma$	4.5 min	1.5 spect.	0.37	Gd- $n$ - $\gamma$
65	Tb <sup>152</sup> Tb <sup>153</sup>	D D		K K, $e^-$	4.5 hr 5.1 days	0.15, 0.4( $e^-$ ) abs. Al		Eu- $\alpha$ -3 $n$ Eu- $\alpha$ -2 $n$
	Tb <sup>154</sup>	D		$\beta^+$ , K, $\gamma$ , $e^-$	17.2 hr	2.6( $\beta^+$ ), 0.22, $\sim 1$ ( $e^-$ ) spect., abs. Al 0.1( $e^-$ ) abs. Al	1.4 abs. Pb	Eu- $\alpha$ -3 $n$ Gd- $p$ - $n$
	Tb <sup>156</sup>	D		K, $e^-$	$\sim 1$ yr			Eu- $\alpha$ -2 $n$
	Tb <sup>159</sup> Tb <sup>160</sup> Tb <sup>160</sup>	A A m.s.	100	$\beta^-$ $\beta^-$ , $\gamma$	3.9 hr 73.5 days; 77.3 days	0.546, 0.882 spect.; 0.75 abs. Al; 0.71 abs. Al	0.086, 0.195, 0.212, 0.297, 1.15 spect. conv., abs. Pb	Tb- $n$ - $\gamma$ Gd- $d$ -2 $n$ Tb- $n$ - $\gamma$
	Tb <sup>161</sup> Tb <sup>161</sup>	F B		$\beta^-$ , $\gamma$ $\beta^-$ , $\gamma$	420 days 5.5 days	0.23 0.5 abs. Al	$\sim 0.1$ , 0.5 1.28 abs. Pb	U- $n$ Gd- $d$ - $n$
66	Dy <sup>156</sup> Dy	F	0.0524	$\beta^+$	2.2 min			Dy- $n$ - $\gamma$ (?)

Isotope Z     A	Class	Per cent abundance	Type of radiation	Half-life	Energy of radiation, Mev		Produced by
					Particles	$\gamma$ -rays	
66 Dy <sup>158</sup> Dy <sup>160</sup> Dy <sup>161</sup> Dy <sup>162</sup> Dy <sup>163</sup> Dy <sup>164</sup> Dy <sup>165m</sup>  Dy <sup>165</sup>	        A res.n.act. A m.s.	        0.0902 2.294 18.88 25.53 24.97 28.18	        I.T., $e^-$  $\beta^-, \gamma$	        1.25 min  145 min; 140 min; 2.5 hr	        0.13( $e^-$ ) abs. A1 0.42, 0.88, 1.25 spect.; 1.20 abs. coincid.; 1.18 spect.; 1.40 cl.ch.	        0.091, 0.37, 0.78 spect. conv., spect.; 1.1 abs. coincid.; ~ 1, 0.37 spect.	        Dy- $n-\gamma$ Dy <sup>164</sup> - $n-\gamma$ Dy- $n-\gamma$ Dy <sup>164</sup> - $n-\gamma$
67 Ho <sup>160</sup> Ho <sup>161, 162</sup>   Ho <sup>162, 161</sup>   Ho <sup>163</sup>  Ho <sup>164</sup>  Ho <sup>166</sup> Ho <sup>166</sup>	 D C   C   B D  A m.s.	          100	 K(?) K(?), $\gamma, e^-$   $\beta^+, K, \gamma$   K, $e^-$ $\beta^-$  $\beta^-$	 ~ 20 min 60 days   4.5 hr   7 days 35 min; 47 min 27.0 hr; 27.5 hr; 27.3 hr; 30 hr	 0.6, 0.16( $e^-$ ) abs. A1   2.0( $\beta^+$ ), 0.3( $e^-$ ) spect., abs. A1 0.4( $e^-$ ) abs. A1 0.7 abs. A1 1.8 abs. A1; 1.9 abs.; 1.6 abs.	             1.1 abs. Pb	             Tb- $\alpha$ -3 $n$ Tb- $\alpha$ -2 $n$ Dy- $d$ -2 $n$ Dy- $p$ - $n$ Tb- $\alpha$ - $n$ Dy- $p$ - $n$   Dy- $p$ - $n$  Ho- $n$ -2 $n$ (?) Dy- $p$ - $n$  Ho- $n$ - $\gamma$
68 Er <sup>162</sup> Er <sup>164</sup> Er <sup>166</sup> Er <sup>166</sup> Er <sup>167</sup> Er <sup>168</sup> Er <sup>169</sup> Er <sup>169, 171</sup>  Er <sup>170</sup> Er <sup>171</sup>   Er <sup>171</sup>	       B F   B  B  F	             0.1 1.5  32.9 24.4 26.9   14.2	             $\beta^+$  $\beta^-$  $\beta^-, \gamma, e^-$  $\beta^-, \gamma$	             1.1 min  9.4 days 6 min; 7 min  7.5 hr; 5.7-7.1 hr; 12 hr  20 hr	             0.33 spect.   1.49(6 %), 1.05 (71 %), 0.67 (22 %) spect., coincid. 0.6 abs. A1	             No $\gamma$   0.81(22 %), 0.31 (71 %), 0.113 (71 %) spect., spect., conv.	             Er- $n$ -2 $n$ (?)     Er- $n$ - $\gamma$ Er- $n$ - $\gamma$   Er- $n$ - $\gamma$ , parent of Tm <sup>171m</sup> (70 %), Tm <sup>171</sup> (30 %)  Er- $n$ - $\gamma$

Isotope Z A	Class	Per cent abundance	Type of radiation	Half-life	Energy of radiation, Mev		Produced by
					Particles	$\gamma$ -rays	
69 Tm <sup>164</sup>	B		$\beta^+$ , K, $\gamma$ , $e^-$	7.7 hr	2.1 ( $\beta^+$ ), 0.24, $\sim 1(e^-)$ spect., abs. Al	1.5 abs. Pb	Ho- $\alpha$ -3n
Tm <sup>167</sup>	B		K, $\gamma$ , $e^-$	9 days	0.21 ( $e^-$ ) abs. Al	0.22, 0.95 abs. Pb	Ho- $\alpha$ -2n Ta-d-5s16a Ho- $\alpha$ -2n
Tm <sup>167,168</sup>	C		K(?), $e^-$	$\sim 100$ days	0.16, 0.5 ( $e^-$ ?) abs. Al		
Tm <sup>169m</sup>	B		I.T., $\gamma$ , $e^-$	$1 \times 10^{-6}$ sec	0.12 ( $e^-$ ) coincid. abs.		Yb <sup>169</sup> K-decay
Tm <sup>169</sup> Tm <sup>170</sup>	A	100	$\beta^-$ , $\gamma$	127 days; $\sim 125$ days; 105 days	0.98 spect.; 1.1 abs. Al	0.83 spect. conv., spect.	Tm-d-p Tm-n- $\gamma$
Tm <sup>171m</sup>	B		I.T., $e^-$	$2.5 \times 10^{-6}$ sec		0.113 spect. conv.; 0.1 coincid. abs. of $e^-$	Er <sup>171</sup> (7.5 hr) $\beta^-$ -decay
Tm <sup>171</sup>	B		$\beta^-$	500 days	0.1 abs. Al; 0.100 spect.		Er <sup>171</sup> (7.5 hr) $\beta^-$ -decay
70 Yb <sup>168</sup> Yb <sup>169</sup>	B	0.06	K, $\gamma$ , $e^-$ (?)	33 days; 33.5 days; 32.5 days		0.2, 0.4 abs. Pb, co- incid.	Tm-d-2n Yb-n- $\gamma$
Yb <sup>170</sup>		4.21					
Yb <sup>171</sup>		14.26					
Yb <sup>172</sup>		21.49					
Yb <sup>173</sup>		17.02					
Yb <sup>174</sup>		29.58					
Yb <sup>175</sup>	A m.s.		$\beta^-$ , $\gamma$	99 hr; 100 hr; 102 hr	0.50, 0.13 abs. Al; 0.45 cl.ch.	0.35 abs. Pb, co- incid.	Yb-n- $\gamma$
Yb <sup>176</sup> Yb <sup>177</sup>	B	13.38	$\beta^-$	2.4 hr; 2.7 hr; 3.5 hr; 1.9 hr	1.3; 1.15 cl.ch.		Yb-n- $\gamma$
71 Lu <sup>170</sup>	B		K, $\gamma$ , $e^-$ , $\beta^+$	2.15 days	1.7 ( $\beta^+$ ), 0.1 ( $e^-$ ) spect., abs. Al	1.5 abs. Pb	Tm- $\alpha$ -3n Yb-d-2n Ta-d-3s13a
Lu <sup>171</sup>	B		K(?), $\gamma$ , $e^-$	9 days	0.17, 0.7 ( $e^-$ ) abs. Al		Tm- $\alpha$ -2n Yb-d-2n Ta-d-3s12a Tm- $\alpha$ -2n Yb-d-2n
Lu <sup>171,172</sup>	C			$> 100$ days	0.11, 0.22 ( $e^-$ ?) abs. Al		

Isotope Z A	Class	Per cent abundance	Type of radiation	Half-life	Energy of radiation, Mev		Produced by
					Particles	$\gamma$ -rays	
71 Lu <sup>175</sup> Lu <sup>176</sup>	A	97.5 2.5	$\beta^-$ , (33 %), $\gamma$ ; K(67 %)	$7.3 \times 10^{10}$ yr (uncorr. for K); $2.4 \times 10^{10}$ yr (corr. for K)	0.215 abs. Al, spect.; 0.40 abs. Al	0.260 abs. Pb	Natural source
Lu <sup>176m</sup>	B		$\beta^-$	3.67 hr; 3.75 hr; 3.7 hr; 3.4 hr	1.04 abs. Al; 1.15 abs. Al; 1.25 cl.ch.	No $\gamma$	Lu-d- $\beta$ Lu-n- $\gamma$ Lu-x-rays
Lu <sup>177</sup>	A m.s.		$\beta^-$ , $\gamma$	6.8 days; 6.6 days; 6.9 days	0.440 abs. Al; 0.52 abs. Al; 0.47 cl.ch.	0.2 abs. Pb; 0.2, 1.3 (weak) abs. Pb	Lu-n- $\gamma$ Lu-d- $\beta$ Hf-d- $\alpha$
72 Hf <sup>174</sup> Hf <sup>175</sup>	B	0.18	K, $\gamma$ , $e^-$	70 days	0.3( $e^-$ ) abs. Al	0.3, 1.5 abs. Pb	Lu-d-2 $\pi$ Lu-p- $\pi$
Hf <sup>174</sup> Hf <sup>177</sup> Hf <sup>178</sup> Hf <sup>179</sup> Hf <sup>180</sup> Hf <sup>181</sup>	A	5.30 18.47 27.10 13.84 35.11	$\beta^-$ , $\gamma$	46 days; 55 days	0.460 spect.; 0.45 abs. Al; 0.42 abs. Al; 0.28 abs. Al, co- incid.; 0.63 abs., coincid.	0.485, 0.347, 0.134, 0.087 spect. conv.; 0.342, 0.128, 0.472 spect. conv., coincid.; 0.52, 0.30 abs. Pb; 1.4 coin- cid. abs.; 0.52, 0.13 abs., coin- cid. abs.	Hf-n- $\gamma$ , parent of Ta <sup>181m</sup>
Hf <sup>m</sup>	D		I.T., $e^-$ (?)	19 sec	0.19( $e^-$ ) abs. Al		Hf-n- $\gamma$
73 Ta <sup>176</sup>	B		K, $\gamma$ , $e^-$	8.0 hr	0.12, 0.18, 1.2( $e^-$ ) abs. Al	1.7 abs. Pb	Lu- $\alpha$ -3 $\pi$ Ta-d-p6 $\pi$
Ta <sup>177</sup>	B		K, $e^-$	2.66 days	0.1( $e^-$ ) abs. Al		Lu- $\alpha$ -2 $\pi$ Hf-d-2 $\pi$ Ta-d-p5 $\pi$
Ta <sup>178, 177</sup>	C		K, $e^-$ , or $\beta^-$	16 days	1.1( $e^-$ ?) abs. Al		Lu- $\alpha$ - $\pi$ Hf-d-2 $\pi$

Isotope Z    A	Class	Per cent abundance	Type of radiation	Half-life	Energy of radiation, Mev		Produced by
					Particles	$\gamma$ -rays	
73 Ta <sup>180</sup>	A		K, $e^-$ , $\gamma$ ; $\beta^-$ (?)	8.2 hr	< 0.5( $e^-$ ?) abs.		Ta-n-2n
Ta <sup>181m</sup>	A		I.T., $\gamma$ , $e^-$	$2.0 \times 10^{-8}$ sec; $2.2 \times 10^{-5}$ sec	0.12( $e^-$ ) coincid. abs.	0.128, 0.472 spect. conv., coincid.; 0.20, 0.49 coincid. abs.	Ta- $\gamma$ -n Hf <sup>181</sup> $\beta^-$ -decay
Ta <sup>181</sup> Ta <sup>182</sup>	A	100	$\beta^-$ , $\gamma$ , $e^-$	117 days; 113 days	1.0 abs.; 0.98, 0.32, 0.050; 0.53 spect.; 0.499 spect.; 1.1 abs. Al, coincid.	1.22(57 %), 1.13 (37 %), 0.22(4 %), 0.15(2 %), spect., conv., 1.6(ZZ); 0.23 abs. Pb	Ta-n- $\gamma$ Ta-d-p
Ta <sup>182m</sup>	E		I.T.(?)	0.40 sec			Ta-n- $\gamma$
Ta <sup>182</sup>	B		$\beta^-$ , $\gamma$ (?)	16.2 min	0.2 abs. Al		Ta-n- $\gamma$
74 W <sup>179, 178</sup>	C		K, $e^-$ , $\gamma$	135 min	0.15, 0.45( $e^-$ ) abs. Al	$\sim$ 0.5, 1.2 abs. Pb	Ta-d-4n
W <sup>180</sup> W <sup>181</sup>	0.122 B		K, $\gamma$ , $e^-$	140 days		$\sim$ 0.14, 1.83(weak) abs. of $e^-$ , abs. Pb	Ta-d-2n
W <sup>182</sup> W <sup>183</sup> W <sup>184</sup> W <sup>185</sup>	A	25.77 14.24 30.68	$\beta^-$ , $\gamma$ (?)	73.2 days; 74 days; 77 days	0.428 spect.; 0.430 spect.; 0.6 abs. Al	No $\gamma$	W-n- $\gamma$ W-n-2n W-d-p Re-d- $\alpha$
W <sup>186</sup> W <sup>m</sup>	D	29.17	I.T., $e^-$	5.5 sec	$\sim$ 0.080( $e^-$ ) abs. Al		W-n- $\gamma$
W <sup>187</sup>	A		$\beta^-$ , $\gamma$ , $e^-$	24.1 hr	0.63(70 %), 1.33(30 %) spect.; 0.562, 1.35 spect.; 1.3, 0.6 spect.; 1.4, 0.6 abs. Al	0.135, 0.101, 0.086 spect. conv.; 0.135, 0.48, 0.69 spect. conv.; 0.90 coin- cid. abs.,	W-n- $\gamma$ W-d-p U- $\alpha$ -55a20s

Isotope $Z$ $A$	Class	Per cent abundance	Type of radiation	Half-life	Energy of radiation, Mev		Produced by
					Particles	$\gamma$ -rays	
74 W <sup>187</sup>						coincid.; 0.14, 0.21, 0.48, 0.62, 0.69 spect. conv.	
75 Re	E		$\beta^+$	30-55 min			W-p-n
Re	E			13 min			W-p-n
Re <sup>182</sup>	B		K, $\gamma$ , $e^-$	64 hr	0.11, 0.27, 0.6( $e^-$ ) abs. Al	0.22, 1.52 abs. Pb	Ta- $\alpha$ -3n W-p-n
Re <sup>183, 184</sup>	C		K, (?), $e^-$	13 hr		1.6 abs. Pb	W-p-2n
Re <sup>184, 183</sup>	C		K, $\gamma$ , $e^-$	$\sim 80$ days	0.1( $e^-$ ) abs. Al	1.0 abs. Pb	Ta- $\alpha$ -n Ta- $\alpha$ -n
Re <sup>184</sup>	A		$\beta^-$ , K, $\gamma$	50 days; 52 days	0.22-0.26; 0.1( $e^-$ ), 0.22, 0.86( $e^-$ ?) abs. Al	0.17, 1.05; 0.85; 0.17, 1 spect. conv., abs. Pb	W-p-n W-d-n Re-n-2n
Re <sup>185</sup> Re <sup>186</sup>	A m.s.	37.07	$\beta^-$	92.8 hr; 90 hr	1.07 abs. Al; 1.05 cl.ch.	No $\gamma$	W-d-2n W-p-n Re- $\gamma$ -n Re-n- $\gamma$ Re-n-2n Re-d-p W <sup>187</sup> $\beta^-$ -decay
Re <sup>187m</sup>	A		I.T., $e^-$ , $\gamma$	$0.65 \times 10^{-6}$ sec	$\leq 0.13$ ( $e^-$ ) coincid. abs.		
Re <sup>187</sup>		62.93	$\beta^-$	$4 \times 10^{11}$ yr	0.043 abs. Al		Natural source
Re <sup>188</sup>	A m.s.		$\beta^-$ , $\gamma$ , $e^-$	18.9 hr; 18 hr	2.05 abs. Al; 2.5 cl.ch. (K.U.); 2.5 abs.; 0.12 ( $e^-$ ), 0.23 ( $\beta^-$ ) coincid. abs.	0.16, 0.48, 0.64, 0.94, 1.43 spect.; 0.7 abs. Pb; 1.39 coin- cid. abs., coincid.	Re-n- $\gamma$ Re-d-p U- $\alpha$ -19s54a
76 Os <sup>184</sup> Os <sup>185</sup>	B	0.018	K, $\gamma$	97 days; 94.7 days		0.75 abs. Pb	Re-d-2n Os-n- $\gamma$
Os <sup>186</sup> Os <sup>187</sup> Os <sup>188</sup> Os <sup>189</sup> Os <sup>190</sup> Os <sup>191</sup>	B	1.59 1.64 13.3 16.1 26.4	$\beta^-$ , $\gamma$ , $e^-$	15.0 days; 16.1 days; 17 days	0.142 spect.; $\leq 0.16$ abs. Al;	0.039, 0.127 spect. conv.; 0.13 abs.	Os-n- $\gamma$ U- $\alpha$ -18s51a

Isotope Z      A	Class	Per cent abundance	Type of radiation	Half-life	Energy of radiation, Mev		Produced by
					Particles	$\gamma$ -rays	
76 Os <sup>191</sup>					0.35 abs. Al; 0.64 coincid. abs.	Pb; 0.129 spect. conv.	
Os <sup>192</sup> Os <sup>192</sup>	A	41.0	$\beta^-$ , $\gamma$	32 hr; 31.9 hr; 30 hr	1.5 abs. Al; 0.95 abs. Al; 1.15 abs. Al; 0.14( $e^-$ ) coincid. abs.	1.17 abs. Pb; 1.58 coincid. abs.	Os-n- $\gamma$ Os-d- $p$ Ir-d-2 $p$ (?)
77 Ir <sup>190</sup>	B		K(?), $e^-$ (?), $\gamma$	10.7 days	0.091( $e^-$ ) abs. Al	0.25 abs. Pb	Os-d-n Ir-n-2n Ir- $\gamma$ -n
Ir <sup>191</sup> Ir <sup>192m</sup>	A res.n.act.	38.5	I.T., $\gamma$ , $e^-$	1.5 min	0.038( $e^-$ ) abs. Al	0.06 abs. Al of $e^-$ , abs. Pb	Ir-n- $\gamma$
Ir <sup>192</sup>	A m.s.		$\beta^-$ , $\gamma$ , $e^-$ ( $\sim 30\%$ )	70 days; 60 days; 75 days	0.67 spect.; 0.68 coin- cid. abs.; 0.56 abs. Al; 0.59 abs. Al	0.307, 0.467, 0.603 spect.; 0.137, 0.209, 0.295, 0.307, 0.316, 0.468, 0.488, 0.591, 0.607, 0.615 spect. conv.; 0.52 abs. Pb	Os-d-2n Ir-n- $\gamma$ Ir-n-2n Ir-d- $p$ Ir- $\gamma$ -n
Ir <sup>193</sup> Ir <sup>194</sup>	A m.s.	61.5	$\beta^-$ , $\gamma$	19.0 hr; 19 hr; 20.7 hr	2.2 spect.; 2.18 spect.; 2.11 abs. Al; 2.07 abs. Al; 0.48 coin- cid. abs.	1.35 spect.; 1.65, 0.38 abs. Pb; coincid.	Ir-n- $\gamma$ Au-d- $\alpha p$ (?) Ir-d- $p$
78 Pt <sup>191</sup>	B		K, $e^-$ , $\gamma$	3.00 days	0.5( $e^-$ ) abs. Al	0.57, 1.5 abs. Pb	Pt-n-2n Ir-d-2n Au <sup>191</sup> K- or $\beta^+$ -decay
Pt <sup>192</sup>		0.78					



Isotope Z A	Class	Per cent abundance	Type of radiation	Half-life	Energy of radiation, Mev		Produced by
					Particles	$\gamma$ -rays	
78 Pt <sup>192</sup>	B		K, $\gamma$ , $e^-$	4.33 days	0.11( $e^-$ ) abs. Al; 0.115( $e^-$ ) abs. Al	0.18, 1.5 abs. of $e^-$ , abs. Pb	Ir- $\alpha$ - $p_n$ Ir- $d$ -2 $n$ Pt- $n$ - $\gamma$ Pt- $d$ - $p$ Pt- $n$ -2 $n$ Au <sup>192</sup> K-decay
Pt <sup>194</sup>		32.8					
Pt <sup>196</sup>		33.7					
Pt <sup>196m</sup>	D	25.4	I.T., $e^-$ , $\gamma$	80 min		0.337 spect. conv.	Pt- $d$ - $p$ Hg- $n$ - $\alpha$ Pt- $n$ - $\gamma$ Pt- $d$ - $p$ Pt- $n$ -2 $n$ Hg- $n$ - $\alpha$ Pt- $n$ - $\gamma$ Pt- $d$ - $p$
Pt <sup>197</sup>	B		$\beta^-$	18 hr	0.65 abs.; 0.72 abs.		
Pt <sup>197</sup>	B		$\beta^-$ , $\gamma$	3.3 days			
Pt <sup>198</sup>		7.23					
Pt <sup>199</sup>	A		$\beta^-$	31 min	1.8 abs.		Pt- $n$ - $\gamma$ Pt- $d$ - $p$ Hg- $n$ - $\alpha$
79 Au <sup>&lt;190</sup>	F		$\alpha$	$\sim 5$ min	5.2 ion. ch.		Au- $d$ -?
Au <sup>191</sup>	D		K or $\beta^+$	$\sim 1$ day			Ir- $\alpha$ -4 $n$ Pt- $d$ -3 $n$ , parent of Pt <sup>191</sup>
Au <sup>192</sup>	B		K, $\gamma$ , $e^-$	4.7 hr	0.4( $e^-$ ) abs. Al	2.3 abs. Pb	Ir- $\alpha$ -3 $n$ Pt- $d$ -2 $n$
Au <sup>192</sup>	B		K, $e^-$	15.8 hr	$< 0.2$ ( $e^-$ ) abs. Al		Ir- $\alpha$ -2 $n$ Pt- $d$ -3 $n$ Parent of Pt <sup>192</sup>
Au <sup>194</sup>	B		K, $\gamma$ , $e^-$	39.5 hr; 39 hr	0.31, 1.8( $e^-$ ) abs. Al	0.329, 1.48, 0.286, 0.46, 2.0 spect. conv., spect.; 0.4, 1.8 abs. of $e^-$ , abs. Pb	Ir- $\alpha$ -3 $n$ Pt- $d$ -2 $n$ Pt- $p$ - $n$
Au <sup>196</sup>	B		K, $\gamma$ , $e^-$	185 days; 180 days	0.1( $e^-$ ) abs. Al	0.19, 1.6 abs. of $e^-$ , abs. Pb; 0.096 (90 %), 0.129 (10 %) spect. conv., coincid.	Ir- $\alpha$ -2 $n$ Pt- $d$ -2 $n$ Pt- $p$ - $n$
Au <sup>196</sup>	B		$\beta^-$ ; K or I.T.	14.0 hr; 13 hr			Au- $n$ -2 $n$

Isotope $Z$ $A$	Class	Per cent abundance	Type of radiation	Half-life	Energy of radiation, Mev		Produced by
					Particles	$\gamma$ -rays	
79 Au <sup>196</sup>	B	100	$\beta^-$ , $\gamma$ , $e^-$ K (70 %), $\beta^-$ (30 %)	5.55 days; 5.6 days	$\sim 0.27$ , $\sim 0.43$ spect., coincid.; 0.36	0.139, 0.358 (with K), 0.173, 0.334 (with $\beta^-$ ) spect. conv. coincid.; 0.41; 0.41, 1.7 abs. Pb	Au- $n-2n$ Pt- $d-n$ Pt- $p-n$
Au <sup>197</sup> Au <sup>197m</sup>	A		I.T., $e^-$	7.5 sec	0.07 ( $e^-$ ), 0.25 ( $e^-$ ) abs. Al, coincid.	0.273 spect. conv. coincid.; 0.25 abs. K-decay of $e^-$	Au-x-rays Au- $n-n$ Hg <sup>197</sup> (25 hr) K-decay (4 %)
Au <sup>198</sup>	A		$\beta^-$ , $\gamma$ , $e^-$ (4.7 %)	2.69 days; 2.7 days; 65.5 hr	0.960 (100 %) spect.; 0.970 (85 %), 0.605 (15 %) spect.; 0.97 (100 %) spect.; 0.985 abs. Al, coincid.; coincid.	0.4112 cryst. spect.; 0.408 (100 %), 0.157 (15 %), 0.208 (15 %) spect., spect. conv.; 0.065 abs., coincid.; coincid.	Au- $n-\gamma$ Au- $d-p$ Hg- $n-p$ Pt- $p-n$ U- $\alpha$ -15z44a
Au <sup>199</sup>	A		$\beta^-$ , $\gamma$	3.3 days	0.38 abs. Al, coincid.; 1.01 abs.	0.18 abs. Pb, coin- cid.; 0.45 abs.	Pt <sup>199</sup> $\beta^-$ -decay Pt- $d-n$ Hg- $n-p$ Hg- $n-p$ Tl- $n-\alpha$
Au <sup>200, 202</sup>	D	0.15	$\beta^-$	48 min	2.5 abs.		
80 Hg <sup>&lt;196</sup> Hg <sup>196</sup> Hg <sup>197</sup>	E		$\alpha$	0.7 min	5.7 ion.ch.		Au- $d-?$
	A		K, $\gamma$ , $e^-$	23 hr; 25 hr		0.161, 0.130 spect. conv.; 0.125, 0.157 spect. conv.; 0.165, 0.135 coincid. abs. spect. conv.	Pt- $\alpha-n$ Au- $d-2n$ Au- $p-n$ Hg- $n-2n$ Hg- $n-\gamma$ Hg- $d-p$
Hg <sup>197</sup>	A		K, $\gamma$ , $e^-$	64 hr		0.075 spect. conv.;	Au- $d-2n$ Au- $p-n$

Isotope $Z$ $A$	Class	Per cent abundance	Type of radiation	Half-life	Energy of radiation, Mev		Produced by
					Particles	$\gamma$ -rays	
80 Hg <sup>197</sup>						0.077 abs. of $e^-$ , spect. conv. 0.4 $\beta^-e^-$ coincid.(?)	Hg- $n-2n$ Hg- $n-\gamma$
Hg <sup>198m</sup>	F		I.T., $\gamma$ , $e^-$	$\sim 0.3 \times 10^{-8}$ sec			Au <sup>198</sup> $\beta^-$ -decay
Hg <sup>198</sup> Hg <sup>199</sup> Hg <sup>m</sup>	D	10.1 17.0	I.T., $e^-$ , $\gamma$	43 min; 43.5 min		$\sim 0.53$ abs. of $e^-$ ; 0.222, 0.362 (spect. conv.)	Pt- $\alpha-n$ (?) Hg- $n-2n$ Hg- $n-n$ (?) Hg- $d-p$ Hg-x-rays
Hg <sup>200</sup> Hg <sup>201</sup> Hg <sup>202</sup> Hg <sup>203, 206</sup>	C	23.3 13.2 29.6	$\beta^-$ , $\gamma$ , $e^-$	45.8 days; 51.5 days	0.205 spect.; < 0.3 spect.; 0.46 abs. Al; 0.11, 0.44 coin- cid. abs.	0.286 spect. conv.; 0.30 abs. Pb; 0.28 spect.	Hg- $n-\gamma$ Hg- $d-p$ Tl- $n-p$
Hg <sup>204</sup> Hg <sup>206</sup>	A	6.7	$\beta^-$	5.5 min	1.62 abs. Al		Hg- $d-p$ Hg- $n-\gamma$ Tl- $n-p$ Pb- $n-\alpha$
81 Tl	D		K(?), $e^-$ , $\gamma$	10.5 hr		1.0 abs. Pb	Hg- $d-2n$
Tl	D		K(?), $e^-$	44 hr			Hg- $d-2n$
Tl <sup>196</sup>	D		K, $\gamma$ , $e^-$	1.8 hr	0.4( $e^-$ ) abs. Al, Be	1.3 abs. Pb	Au- $\alpha-3n$
Tl <sup>199</sup>	B		K, $\gamma$ , $e^-$	7 hr; 7.5 hr	0.5( $e^-$ ) abs. Al, Be	1.5 abs. Pb	Au- $\alpha-2n$ Pb <sup>199</sup> K-decay
Tl <sup>200</sup>	B		K, $\gamma$ , $e^-$	27 hr	0.4( $e^-$ ) abs. Al, Be		Au- $\alpha-n$ Pb <sup>200</sup> K-decay
Tl <sup>201</sup>	D		K	75 hr			Pb <sup>201</sup> K-decay
Tl <sup>202</sup>	B		K(?), $\gamma$ , $e^-$	11.8 days; 13 days		0.40	Hg- $d-2n$ Tl- $n-2n$
Tl <sup>203</sup> Tl <sup>204</sup>	B	29.1	$\beta^-$	2.7 yr; 3.5 yr	0.80 abs. Al; 0.87 cl.ch.; 0.77 spect.	No $\gamma$	Tl- $n-\gamma$ Tl- $d-p$
Tl <sup>206</sup> Tl <sup>206</sup>	A	70.9	$\beta^-$	4.23 min	1.65 abs.; 1.77 abs. Al	No $\gamma$	Tl- $n-\gamma$ Tl- $d-p$ Pb- $\gamma-p$ RaE <sup>210</sup> $\alpha$ -decay

Isotope Z      A	Class	Per cent abundance	Type of radiation	Half-life	Energy of radiation, Mev		Produced by
					Particles	$\gamma$ -rays	
81 AcC'' <sup>207</sup>	A		$\beta^-$ , $\gamma$	4.76 min	1.47 abs. Al		Pb-n- $p$ Natural source, AcC <sup>211</sup> $\alpha$ -decay
ThC'' <sup>208</sup>	A		$\beta^-$ , $\gamma$	3.1 min	1.72 spect.; 1.82 abs. paper	2.62	Natural source, ThC <sup>212</sup> $\alpha$ -decay
Tl <sup>209</sup>	A		$\beta^-$	2.2 min	1.8 abs. Al		Bi <sup>213</sup> $\alpha$ -de- cay, parent of Pb <sup>209</sup>
RaC'' <sup>210</sup>	A		$\beta^-$	1.32 min	1.80 cl.ch.		Natural source, RaC <sup>214</sup> $\alpha$ -decay, parent of RaD <sup>210</sup>
82 Pb <sup>199</sup>	B		K	1-2 hr			Bi <sup>199</sup> K-de- cay, par- ent of Tl <sup>199</sup>
Pb <sup>200</sup>	B		K	18 hr.			Bi <sup>200</sup> K-de- cay, par- ent of Tl <sup>200</sup>
Pb <sup>201</sup>	D		K, $e^-$ , $\gamma$	8 hr; $\sim$ 5 hr			Tl-d-4n
Pb <sup>203</sup>	B		I.T.(?) or K(?) $e^-$ , $\gamma$	52 hr; 54 hr		$\sim$ 0.45 abs. of $e^-$ , abs. Pb, spect., spect. conv., 0.27 spect. conv., abs. Pb	Tl-d-2n Tl-p-n Pb-n-2n Pb <sup>204</sup> n-2n Pb- $\gamma$ -n
Pb <sup>204</sup> Pb <sup>204m</sup>	B	1.5	I.T., $\gamma$ , $e^-$	68 min; 65 min		1.1 abs. of $e^-$ , abs. Pb; 0.90	Tl-d-n Tl-d-3n Pb-n-n Pb-x-rays Bi <sup>204</sup> K-decay
Pb <sup>206</sup> Pb <sup>207</sup> Pb <sup>208</sup> Pb <sup>209</sup>	A	23.6 22.6 52.3	$\beta^-$	3.32 hr	0.70 abs.; 0.68 spect.; 0.750; 0.71 abs. Al; 0.70 spect.	No $\gamma$ , no $e^-$ ; no $\gamma$	Pb-d-p Pb-n- $\gamma$ Bi-n-p Po <sup>212</sup> $\alpha$ -decay
RaD <sup>210</sup>	A		$\beta^-$ , $\gamma$	22 yr	0.0255 spect.; 0.0292	0.047; 0.0472 spect.	Natural source, RaC'' <sup>210</sup>

Isotope $Z$ $A$	Class	Per cent abundance	Type of radiation	Half-life	Energy of radiation, Mev		Produced by
					Particles	$\gamma$ -rays	
82 $\text{RaD}^{210}$					spect.	conv.; several weak lines of lower energy	$\beta^-$ -decay, $\text{RaC}^{214}$ $\alpha$ -decay, parent of $\text{RaE}^{210}$
$\text{AcB}^{211}$	A		$\beta^-, \gamma$	36.1 min	0.5, 1.40 abs. Al	0.8 abs.	Natural source, $\text{AcA}^{215}$ $\alpha$ -decay, parent of $\text{AcC}^{211}$
$\text{ThB}^{212}$	A		$\beta^-, \gamma$	10.6 hr	0.36 spect.		Natural source, $\text{ThA}^{216}$ $\alpha$ -decay, parent of $\text{ThC}^{212}$
$\text{RaB}^{214}$	A		$\beta^-, \gamma$	26.8 min	0.65 spect.		Natural source, $\text{RaA}^{218}$ $\alpha$ -decay, parent of $\text{RaC}^{214}$
83 $\text{Bi}^{197}$	E		$\alpha$	2 min	6.2 ion.ch.		$\text{Pb-d-?}$
$\text{Bi}^{198}$	D		$\alpha$ ; K(?)	9 min	5.83 ion.ch.; ~ 5.5( $\alpha$ ) ion.ch.		$\text{Pb-d-?}$
$\text{Bi}^{199}$	B		$\alpha$ ; K	27 min	5.47 ion.ch., abs. mica; ~ 5.5( $\alpha$ ) ion.ch.		$\text{Pb-d-?}$
$\text{Bi}^{200}$	B		$\alpha$ ; K	62 min; ~ 100 min	5.15 ion.ch.; ~ 5.5( $\alpha$ ) ion.ch.		$\text{Pb-d-?}$
$\text{Bi}^{204}$	B		K, $e^-, \gamma$	12 hr	0.2( $e^-$ ), ~ 0.8( $e^-$ , weak) spect., abs. Al		$\text{Pb}^{204}\text{-d-}2\pi$ $\text{Ti-}\alpha\text{-}3\pi$ , parent of $\text{Pb}^{204m}$ (~ 4 %)
$\text{Bi}^{206}$	A		K(?), $e^-, \gamma$	6.4 days		0.74 abs. of $e^-$ ; 0.93 abs. of $e^-$ ; 1.1 abs. Pb; ~ 0.4, 1.1 abs. Pb	$\text{Ti-}\alpha\text{-}3\pi$ $\text{Pb-d-}2\pi$ $\text{Pb}^{207}\text{-d-}3\pi$ $\text{Po}^{206}$ K-decay
$\text{Bi}^{208}$	F		K	Short			Bi-n- $2\pi$
$\text{Bi}^{209}$		100					
$\text{RaE}^{210}$	A		$\beta^-$ (~100 %); $\alpha$ ( $10^{-4}$ $10^{-5}$ %)	5.0 days	1.17( $\beta^-$ ) spect.; 4.77( $\alpha$ ) calc.	No $\gamma$	Natural source, $\text{RaD}^{210}$ $\beta^-$ -decay, parent of $\text{Po}^{210}$ and $\text{Ti}^{208}$

Isotope Z      A	Class	Per cent abundance	Type of radiation	Half-life	Energy of radiation, Mev		Produced by
					Particles	$\gamma$ -rays	
83 RaE <sup>210</sup>							Bi- $\alpha$ - $p$ Pb- $\alpha$ - $pn$ Bi- $n$ - $\gamma$ Natural source, AcB <sup>211</sup> $\beta^-$ -decay, parent of AcC' <sup>211</sup> and AcC'' <sup>207</sup> At <sup>218</sup>
AcC <sup>211</sup>	A		$\alpha$ (99.68 %) $\gamma$ ; $\beta^-$ (0.32 %), $\gamma$	2.16 min	6.619 ( $\alpha$ , 84 %), 6.273 ( $\alpha$ , 16 %) spect.		
ThC <sup>212</sup>	A		$\alpha$ (33.7 %), $\gamma$ ; $\beta^-$ (66.3 %), $\gamma$	60.5 min	6.081 ( $\alpha$ , 27 %), 6.042 ( $\alpha$ , 70 %) ( $\alpha$ , others, 3 %) spect.; 2.20 ( $\beta^-$ ) spect.		Natural source, ThB <sup>212</sup> $\beta^-$ -decay, parent of ThC' <sup>212</sup> and ThC'' <sup>208</sup> At <sup>218</sup>
Bj <sup>213</sup>	A		$\beta^-$ ; $\alpha$ (2 %), (4 %)	47 min; 46 min	$\sim 1.3$ ( $\beta^-$ ) abs. Al; $\sim 1.2$ ( $\beta^-$ ); 5.86 ( $\alpha$ ) ion.ch.; 6.0 ( $\alpha$ ) ion.ch.		$\alpha$ -decay, At <sup>217</sup> $\alpha$ -decay, parent of Po <sup>213</sup>
RaC <sup>214</sup>	A		$\alpha$ (0.04 %); $\beta^-$ (99.96 %), $\gamma$	19.7 min	5.505 ( $\alpha$ , 45 %), 5.444 ( $\alpha$ , 55 %) spect.; 3.15 ( $\beta^-$ ) abs. Al, spect.	1.8	Natural source RaB <sup>214</sup> $\beta^-$ -decay, At <sup>218</sup> $\alpha$ -decay, parent of RaC' <sup>214</sup> and RaC'' <sup>210</sup>
84 Po <sup>203</sup>	D		$\alpha$ ; K	40 min	5.56 ( $\alpha$ ) ion.ch.		Pb- $\alpha$ -7n
Po <sup>205</sup>	D		$\alpha$ ; K	4 hr	5.35 ( $\alpha$ ) ion.ch.		Pb- $\alpha$ -5n
Po <sup>206</sup>	A		K ( $\sim 90$ %), $\gamma$ , $e^-$ ; $\alpha$ ( $\sim 10$ %)	9 days	5.2 ( $\alpha$ ) ion.ch.	0.8 abs. Pb	Pb <sup>206</sup> - $\alpha$ -2n, parent of Bi <sup>206</sup>
Po <sup>207</sup>	A		K ( $\sim 100$ %), $\gamma$ ; $\alpha$ (0.01 %)	5.7 hr	5.1 ( $\alpha$ ) ion.ch.	1.3 abs. Pb	Pb <sup>206</sup> - $\alpha$ -3n
Po <sup>208</sup>	B		$\alpha$	3 yr	5.14 ion.ch.	No $\gamma$	Pb <sup>206</sup> - $\alpha$ -2n Pb <sup>207</sup> - $\alpha$ -3n Bi- $d$ -3n Bi- $p$ -2n

Isotope Z      A	Class	Per cent abundance	Type of radiation	Half-life	Energy of radiation, Mev		Produced by
					Particles	$\gamma$ -rays	
84 Po <sup>210</sup>	A		$\alpha, \gamma$	138 days; 140 days	5.298 spect.; 5.303 spect.	0.773 spect. conv.; 0.8 (weak) abs. Pb	Natural source, RaE <sup>210</sup> $\beta^-$ -decay Pb- $\alpha$ -2 $\pi$ Bi-d- $\pi$ At <sup>210</sup> K-decay
AcC <sup>211</sup>	A		$\alpha$	$5 \times 10^{-3}$ sec	7.434 spect.		Natural source, AcC <sup>211</sup> $\beta^-$ -decay At <sup>211</sup> K-decay
ThC <sup>212</sup>	A		$\alpha$	$3.0 \times 10^{-7}$ sec; $3.4 \times 10^{-7}$ sec; $2.6 \times 10^{-7}$ sec; $3 \times 10^{-7}$ sec	8.776 spect.		Natural source, ThC <sup>212</sup> $\beta^-$ -decay Em <sup>212</sup> $\beta^-$ -decay Bi <sup>212</sup> $\beta^-$ -decay, parent of Pb <sup>209</sup> Em <sup>217</sup>
Po <sup>213</sup>	A		$\alpha$	$4.2 \times 10^{-8}$ sec	8.336 ion. ch.; 8.30 ion.ch.		$\alpha$ -decay Natural source, RaC <sup>214</sup> $\beta^-$ -decay, parent of RaD <sup>210</sup> Em <sup>218</sup>
RaC <sup>214</sup>	A		$\alpha$	$1.5 \times 10^{-4}$ sec; $1.55 \times 10^{-4}$ sec; $1.4 \times 10^{-4}$ sec	7.680 spect.		$\alpha$ -decay Natural source, An <sup>213</sup> $\alpha$ -decay, parent of AcB <sup>211</sup> and At <sup>215</sup>
AcA <sup>215</sup>	A		$\alpha$ (~ 100 %); $\beta^-$ ( $5 \times 10^{-4}$ %)	$1.83 \times 10^{-3}$ sec	7.365 spect.		Natural source, Th <sup>230</sup> $\alpha$ -decay, parent of ThB <sup>212</sup> and At <sup>215</sup>
ThA <sup>216</sup>	A		$\alpha$ (~ 100 %); $\beta^-$ (0.014 %)	0.158 sec	6.774 ( $\alpha$ ) spect.		Natural source, Rn <sup>222</sup> $\alpha$ -decay, parent of RaB <sup>214</sup> and At <sup>215</sup>
RaA <sup>226</sup>	A		$\alpha$ (99.96 %); $\beta^-$ (0.04 %)	3.05 min	5.998 ( $\alpha$ ) spect.		Natural source, Rn <sup>222</sup> $\alpha$ -decay, parent of RaB <sup>214</sup> and At <sup>215</sup>

Isotope Z     A	Class	Per cent abundance	Type of radiation	Half-life	Energy of radiation, Mev		Produced by
					Particles	$\gamma$ -rays	
85 At <sup>207</sup>	D		$\alpha$ ; K(?)	1.7 hr	5.76( $\alpha$ ) ion. ch.	1.0	Bi- $\alpha$ -6n
At <sup>208</sup>	D		$\alpha$ ; K(?)	4.5 hr	5.66( $\alpha$ ) ion. ch.		Bi- $\alpha$ -4n
At <sup>210</sup>	A		K, $\gamma$	8.3 hr			Bi- $\alpha$ -3n, parent of Po <sup>210</sup>
At <sup>211</sup>	A		$\alpha$ (40 %); K(60 %)	7.5 hr	5.89( $\alpha$ ) ion. ch.; 5.94 ( $\alpha$ ) abs.		Bi- $\alpha$ -2n Th- $\alpha$ 25a7z U- $\alpha$ -31a9z
At <sup>212</sup>	A		$\alpha$	0.25 sec			Bi- $\alpha$ -n
At <sup>214</sup>	B		$\alpha$	Very short	8.78 ion.ch.		Fr <sup>215</sup>
At <sup>216</sup>	A		$\alpha$	$\sim 10^{-4}$ sec; short	8.00 ion.ch.; 8.4 ion.ch.		$\alpha$ -decay Natural source AcA <sup>215</sup> $\beta^-$ -decay, parent of AcC <sup>211</sup> Fr <sup>219</sup>
At <sup>218</sup>	A		$\alpha$	$3 \times 10^{-4}$ sec; $\sim 10^{-3}$ sec; short ( $< 54$ sec)	7.79 ion.ch.; 7.64 ion. ch.		$\alpha^-$ -decay, parent of AcC <sup>211</sup> Natural source, ThA <sup>218</sup> $\beta^-$ -decay, parent of ThC <sup>212</sup> Fr <sup>220</sup>
At <sup>217</sup>	A		$\alpha$	0.018 sec; 0.021 sec	7.02 ion.ch.; 7.00 ion. ch.		Fr <sup>221</sup> $\alpha$ -decay; parent of Bi <sup>213</sup>
At <sup>218</sup>	F		$\alpha$	Several sec(?)	6.72 ion.ch.		Natural source, RaA <sup>218</sup> $\beta^-$ -decay, parent of RaC <sup>214</sup>
86 Em <sup>216</sup>	A		$\alpha$	Very short	8.07 ion.ch.		Ra <sup>220</sup> $\alpha$ -decay, parent of ThC <sup>212</sup>
Em <sup>217</sup>	A		$\alpha$	$\sim 1 \times 10^{-3}$ sec	7.74 ion.ch.		Ra <sup>221</sup> $\alpha$ -decay, parent of Po <sup>213</sup>
Em <sup>218</sup>	A		$\alpha$	0.019 sec	7.12 ion.ch.; 7.1 ion.ch.		Ra <sup>222</sup> $\alpha$ -decay, parent of RaC <sup>214</sup>



Isotope Z A	Class	Per cent abundance	Type of radiation	Half-life	Energy of radiation, Mev		Produced by
					Particles	$\gamma$ -rays	
86 An <sup>219</sup>	A		$\alpha$	3.92 sec	6.824(82 %), (others 18 %) spect.		Natural source, AcX <sup>223</sup> $\alpha$ -decay, parent of AcA <sup>215</sup>
Tn <sup>220</sup>	A		$\alpha$	54.5 sec	6.282 spect.		Natural source, ThX <sup>224</sup> $\alpha$ -decay, parent of ThA <sup>216</sup>
Rn <sup>222</sup>	A		$\alpha$	3.825 days	5.486 spect.		Natural source, Ra <sup>226</sup> $\alpha$ -decay, parent of RaA <sup>218</sup>
87 Fr <sup>218</sup>	B		$\alpha$	Very short	7.85 ion.ch.		Ac <sup>222</sup> $\alpha$ -decay, parent of At <sup>214</sup>
Fr <sup>219</sup>	A		$\alpha$	$\sim 0.02$ sec	7.30 ion.ch.		Ac <sup>223</sup> $\alpha$ -decay, parent of At <sup>215</sup>
Fr <sup>220</sup>	A		$\alpha$	27.5 sec; $\sim 30$ sec	6.69 ion.ch.		Ac <sup>224</sup> $\alpha$ -decay, parent of At <sup>216</sup>
Fr <sup>221</sup>	A		$\alpha$	4.8 min; 5 min	6.30 ion.ch.		Ac <sup>225</sup> $\alpha$ -decay, parent of At <sup>217</sup>
Fr <sup>223</sup> (AcK)	A		$\beta^-$ , $\gamma$	21 min	1.20 cl.ch.	0.090 abs. Al	Natural source, Ac <sup>227</sup> $\alpha$ -decay, parent of AcX <sup>223</sup>
88 Ra <sup>220</sup>	A		$\alpha$	Short	7.49 ion.ch.		Th <sup>224</sup> $\alpha$ -decay, parent of Em <sup>216</sup>
Ra <sup>221</sup>	A		$\alpha$	31 sec	6.71 ion.ch.		Th <sup>225</sup> $\alpha$ -decay, parent of Em <sup>217</sup>
Ra <sup>222</sup>	A		$\alpha$	38 sec	6.51 ion.ch.; 6.5 ion.ch.		Th <sup>226</sup> $\alpha$ -decay, parent of Em <sup>218</sup>

Isotope Z     A	Class	Per cent abundance	Type of radiation	Half-life	Energy of radiation, Mev		Produced by
					Particles	$\gamma$ -rays	
88 AcX <sup>228</sup>	A		$\alpha, \gamma$	11.2 days	5.717 (55 %), 5.606 (36 %), (others 9 %) spect.		Natural source, RdAc <sup>227</sup> $\alpha$ -decay, AcK <sup>228</sup> $\beta^-$ -decay, Ac <sup>228</sup> K-decay, parent of An <sup>219</sup>
ThX <sup>228</sup>	A		$\alpha$	3.64 days	5.681 spect.; 5.66 ion. ch.		U- $\alpha$ -19a6s U-d-17a5s Natural source, RdTh <sup>228</sup> $\alpha$ -decay, parent of Th <sup>228</sup>
Ra <sup>226</sup>	A		$\beta^-$	14.8 days; 14 days	$\sim 0.2$ abs. Al; < 0.05 abs.		U- $\alpha$ -18a6s U-d-16a5s Ac <sup>224</sup> K-decay Th <sup>229</sup> $\alpha$ -decay, parent of Ac <sup>226</sup>
Ra <sup>226</sup>	A		$\alpha, \gamma$	1622 yr; 1631 yr; 1590 yr	4.791 spect.	0.19	Natural source, Io <sup>230</sup> $\alpha$ -decay, parent of Rn <sup>222</sup>
Ra <sup>227</sup>	A		$\beta^-$				Ra-n- $\gamma$ , parent of Ac <sup>227</sup>
MsTh <sup>228</sup>	A		$\beta^-$	6.7 yr	< 0.015 cl. ch.; 0.053 spect., abs. Al		Natural source, Th <sup>232</sup> $\alpha$ -decay, parent of MsTh <sup>228</sup>
89 Ac <sup>227</sup>	B		$\alpha$	Short	6.96 ion.ch.		Pa <sup>228</sup> $\alpha$ - decay, par- ent of Fr <sup>218</sup>
Ac <sup>228</sup>	A		$\alpha$ (99.9 %); K (0.1 %)	2.2 min	6.64 ion.ch.		Pa <sup>227</sup> $\alpha$ -decay, parent of Fr <sup>219</sup> and AcX <sup>228</sup>
Ac <sup>224</sup>	A		$\alpha$ ( $\sim 10$ %); K ( $\sim 90$ %)	2.9 hr; 2.5 hr	6.17 ion.ch.		Pa <sup>228</sup> $\alpha$ -decay, parent of Fr <sup>220</sup> and ThX <sup>224</sup>

Isotope Z     A	Class	Per cent abundance	Type of radiation	Half-life	Energy of radiation, Mev		Produced by
					Particles	$\gamma$ -rays	
89 Ac <sup>225</sup>	A		$\alpha$	10.0 days	5.80 ion.ch.		Ra <sup>225</sup> $\beta^-$ -decay Th <sup>225</sup> K-decay Pa <sup>229</sup> $\alpha$ -decay, parent of Fr <sup>221</sup> U-d-15a4s U- $\alpha$ -16a5s, parent of Th <sup>225</sup>
Ac <sup>226</sup>	A		$\beta^-$	22 hr			
Ac <sup>227</sup>	A		$\alpha$ (1.2 %), (1.25 %); $\beta^-$ (99 %), $\gamma$ , $e^-$	21.7 yr; 13.5 yr	4.94( $\alpha$ ) (100 %) ion.ch.; 4.95( $\alpha$ ) (85 %), 4.6( $\alpha$ ) (15 %) ion.ch.; 4.95( $\alpha$ ) ion.ch.; < 0.01(?) ( $\beta^-$ )	0.037(weak) abs. Al	Natural source, Pa <sup>231</sup> $\alpha$ -decay, parent of RdAc <sup>227</sup> and AcK <sup>223</sup> Ra <sup>227</sup> $\beta^-$ -decay
MsTh <sub>2</sub> <sup>228</sup>	A		$\beta^-$ , $\gamma$ ; $\alpha$ (?)	6.13 hr	1.55( $\beta^-$ ) spect.; 4.54( $\alpha$ ) abs. air		Natural source, MsTh <sub>2</sub> <sup>228</sup> $\beta^-$ -decay, parent of RdTh <sup>228</sup>
90 Th <sup>224</sup>	A		$\alpha$	Short	7.20 ion.ch.		U <sup>228</sup> $\alpha$ -decay, parent of Ra <sup>220</sup>
Th <sup>225</sup>	A		$\alpha$ (90 %); K(10 %)	7.8 min	6.57 ion.ch.		U <sup>229</sup> $\alpha$ -decay, parent of Ra <sup>221</sup> and Ac <sup>225</sup>
Th <sup>226</sup>	A		$\alpha$	30.9 min	6.30 ion.ch.; 6.3 ion.ch.		U <sup>230</sup> $\alpha$ -decay, parent of Ra <sup>222</sup> Ac <sup>226</sup>
RdAc <sup>227</sup>	A		$\alpha$ , $\gamma$	18.6 days; 18.9 days	6.049(20 %), 5.988 (25 %), 5.764 (20 %), 5.717 (15 %) (others 20 %) spect.		$\beta^-$ -decay Natural source, Ac <sup>227</sup> $\beta^-$ -decay, parent of AcX <sup>223</sup> U-d-13a3s

Isotope Z     A	Class	Per cent abundance	Type of radiation	Half-life	Energy of radiation, Mev		Produced by
					Particles	$\gamma$ -rays	
90 R <sub>d</sub> Th <sup>228</sup>	A		$\alpha, \gamma$	1.90 yr	5.418 (83 %), 5.333 (17 %) spect.; 5.38 ion. ch.		Natural source, MsTh <sub>2</sub> <sup>228</sup> $\beta^-$ -decay, parent of ThX <sub>1</sub> <sup>228</sup> U <sub>232</sub> $\alpha$ -decay Pa <sub>228</sub> K-decay U <sub>233</sub> $\alpha$ -decay, parent of Ra <sub>226</sub>
Th <sup>229</sup>	A		$\alpha$	7000 yr; $\sim 10^4$ yr	5.02, 4.94, 4.85 ion. ch.; 5.05 ( $\sim 10$ %) 4.95 ( $\sim 20$ %), 4.85 ( $\sim 70$ %) ion.ch.		
Io <sup>230</sup>	A		$\alpha, \gamma$	$8.0 \times 10^4$ yr; $8.3 \times 10^4$ yr	4.66 abs. air; 4.81 calor.; 4.66 ion. ch.		Natural source, U <sub>234</sub> <sup>234</sup> $\alpha$ -decay, parent of Ra <sub>226</sub> Pa <sub>230</sub> K-decay Natural source, AcU <sub>235</sub> $\alpha$ -decay, parent of Pa <sub>231</sub>
UY <sub>231</sub>	A		$\beta^-, \gamma, e^-$	25.65 hr; 25.5 hr; 24.0 hr; 24.6 hr	0.21; 0.2 abs.	0.035; 0.035, 0.065 abs. Pb, Cu, Ag	
Th <sup>232</sup>	A	100	$\alpha$	$1.39 \times 10^{10}$ yr	3.98 ion.ch.; 4.20 ion. ch.		Th- $n$ -2 $n$ Natural source, parent of MsTh <sub>1</sub> <sup>232</sup>
Th <sup>232</sup>	A		$\beta^-$	23.5 min; 23 min	1.2 abs. Al	No $\gamma$	Th- $n$ - $\gamma$ Th- $d$ - $p$
UX <sub>1</sub> <sup>234</sup>	A		$\beta^-, \gamma$	24.10 days; 24.1 days; 24.5 days	0.11, 0.20 abs. Al; 0.13 abs Al, spect.; 0.190 spect.; 0.205 spect.	0.092 (1 %) 0.092 spect. conv.; 0.093 (20 %) spect. conv.	Natural source, U <sub>238</sub> $\alpha$ -decay, parent of UX <sub>2</sub> <sup>234m</sup>
91 Pa <sup>226</sup>	B		$\alpha$	1.7 min	6.81 ion.ch.		Th- $d$ -8 $\pi$ , parent of Ac <sub>226</sub>
Pa <sup>227</sup>	A		$\alpha$ ( $\sim 80$ %); K ( $\sim 20$ %)	38 min	6.46 ion.ch.		Th- $d$ -7 $n$ , parent of

Isotope Z     A	Class	Per cent abundance	Type of radiation	Half-life	Energy of radiation, Mev		Produced by
					Particles	$\gamma$ -rays	
91 Pa <sup>227</sup>							Ac <sup>223</sup> and RdAc <sup>227</sup> U- $\alpha$ -3 $\pm$ 15 $\alpha$ Np <sup>221</sup> $\alpha$ -decay Th-d-6n, parent of Ac <sup>224</sup> and RdTh <sup>228</sup>
Pa <sup>228</sup>	A		$\alpha$ (~ 2 %); K(~ 98 %)	22 hr	6.09 ion.ch.		Th <sup>228</sup> -d-3n, parent of Ac <sup>228</sup>
Pa <sup>229</sup>	A		$\alpha$ (~ 1 %); K(~ 99 %)	1.5 days; 1.4 days	5.69 ion.ch.; 5.66 ion. ch.		Th <sup>229</sup> -d-3n, parent of Ac <sup>229</sup>
Pa <sup>230</sup>	A		$\beta^-$ , $\gamma$ ; K (~ 90 %)	17.7 days; 17 days	~ 1.1 abs. Al	0.94 abs. Pb	Parent of U <sup>230</sup> Th- $\alpha$ -p5n Th <sup>230</sup> -d-2n Th-d-4n Pa-d-p2n Pa- $\alpha$ - $\alpha$ n U <sup>231</sup> -d- $\alpha$ n
Pa <sup>231</sup>	A		$\alpha$ , $\gamma$	3.43 $\times 10^4$ yr; 3.2 $\times$ 10 <sup>4</sup> yr	5.012(87 %), 4.736 (13 %) ion.ch.; 5.00 (~ 85 %), 4.69-4.72 (~ 15 %) ion.ch.; 5.049 spect.	0.095, 0.294, 0.323 spect. conv.; 0.308 abs. Pb	Natural source, UY <sup>231</sup> $\beta^-$ -decay, parent of Ac <sup>231</sup> Th-d-3n Th-m-2n, UY <sup>231</sup> $\beta^-$ -decay
Pa <sup>232</sup>	A		$\beta^-$ , $\gamma$ , $e^-$	1.32 days; 1.4 days; 1.6 days	~ 0.28 abs. Al	1.05, ~0.23 abs. Pb; 1.0 abs. Pb	Th-d-2n Th- $\alpha$ -p3n Pa-d-p Th <sup>232</sup>
Pa <sup>233</sup>	A		$\beta^-$ , $\gamma$ , $e^-$	27.4 days	0.4 abs. Al; 0.23 spect.; 0.5 abs. Al; ~ 0.7 spect.	0.084, 0.298, 0.309, 0.337 spect. conv.; $e^-$ lines at 0.063, 0.077, 0.192, 0.293 spect.; 0.33 abs. Pb	$\beta^-$ -decay Np <sup>237</sup> $\alpha$ -decay, parent of U <sup>233</sup> Th-d-n Th- $\alpha$ -p2n
UZ <sup>234</sup>	A		$\beta^-$ , $\gamma$	6.7 hr	0.56, 1.55 abs. Al; 0.45 spect.	0.70 abs. Pb, W	Natural source, UX <sub>2</sub> <sup>234</sup> I.T., parent of U <sub>II</sub> <sup>234</sup>

Isotope Z      A	Class	Per cent abundance	Type of radiation	Half-life	Energy of radiation, Mev		Produced by
					Particles	$\gamma$ -rays	
91 U <sub>X<sub>2</sub></sub> <sup>234m</sup>	A		$\beta^-$ , $\gamma$ ; I.T. (0.15 %)	1.14 min; 1.22 min	1.52 (5 %), 2.32 (95 %) spect.; 2.32 abs. Al	0.802 (5 %) spect. conv.; 0.782, 0.822 spect. conv.; 0.396 (I.T.) spect. conv.	Natural source, U <sub>X<sub>1</sub></sub> <sup>234</sup> $\beta^-$ -decay, parent of U <sub>Z</sub> <sup>234</sup> and U <sub>II</sub> <sup>234</sup>
92 U <sup>228</sup>	A		$\alpha$ (80 %); K (20 %)	9.3 min	6.72 ion.ch.		Th- $\alpha$ -8n, parent of Th <sup>224</sup> Pu <sup>232</sup>
U <sup>229</sup>	A		$\alpha$ (~ 20 %); K (~ 80 %)	58 min	6.42 ion.cl.		$\alpha$ -decay Th- $\alpha$ -7n, parent of Th <sup>225</sup>
U <sup>230</sup>	A		$\alpha$	20.8 days	5.85 ion.ch.; 5.86 ion. ch.		Pa <sup>230</sup> $\beta^-$ -decay, parent of Th <sup>226</sup> Pu <sup>234</sup> $\alpha$ -decay Th- $\alpha$ -6n Pa- $\delta$ -3n Pa- $\alpha$ p4n U- $\delta$ -10as Pa- $\delta$ -2n Pa- $\alpha$ -p3n Th- $\alpha$ -4n Pa <sup>232</sup>
U <sup>231</sup>	B		K	4.2 days			$\beta^-$ -decay Pu <sup>231</sup>
U <sup>232</sup>	A		$\alpha$	70 yr; 30 yr	5.31 abs. Al; 5.27		$\alpha$ -decay, parent of RdTh <sup>228</sup> Pa- $\delta$ -n Pa- $\alpha$ -p2n Pa <sup>233</sup>
U <sup>233</sup>	A		$\alpha$ , $\gamma$ , $e^-$	$1.62 \times 10^5$ yr; $1.63 \times 10^5$ yr; $1.2 \times 10^5$ yr	4.823 ion. ch.; 4.80 abs. air	0.31, 0.080, 0.040 (weak) abs. Pb, Cu, Al	$\beta^-$ -decay, parent of Th <sup>229</sup>
U <sub>II</sub> <sup>234</sup>	A	0.0051	$\alpha$	$2.35 \times 10^5$ yr; $2.69 \times 10^4$ yr	4.763 ion. ch.; 4.78 abs. air; 4.76 ion. ch.		Natural source, U <sub>X<sub>2</sub></sub> <sup>234m</sup> and U <sub>Z</sub> <sup>234</sup> $\beta^-$ -decay, parent of I <sub>o</sub> <sup>230</sup>
AcU <sup>226</sup>	A	0.71	$\alpha$ , $\gamma$	$8.91 \times 10^8$ yr; 7.07	4.56 (20 %), 4.396	0.162 abs. Pb; 0.187	Natural source,

Isotope Z     A	Class	Per cent abundance	Type of radiation	Half-life	Energy of radiation, Mev		Produced by
					Particles	$\gamma$ -rays	
92 AcU <sup>233</sup>				$\times 10^4$ yr; $8.52 \times 10^8$ yr	(80 %) ion.ch.; 4.35; 4.34 ion.ch.	abs. Pb	parent of UY <sup>231</sup>
U <sup>237</sup>	A		$\beta^-$ , $\gamma$ , $e^-$	6.8 days; 6.63 days	$\sim 0.23$ spect.; 0.135, 0.35, (1.6?) abs. Al; 0.26 abs.; 0.17, 0.22 abs. Al, Cellophane	0.057, 0.204, 0.260, 0.032 spect. conv.; 0.14, 0.23, 0.53 abs. Pb	U-n-2n, parent of Np <sup>237</sup> U-d-t U- $\alpha$ - $\alpha$ n Pu <sup>241</sup> $\alpha$ -decay
U <sup>238</sup>	A	99.28	$\alpha$	$4.51 \times 10^9$ yr; 4.498 $\times 10^9$ yr	4.180 ion. ch.; 4.23 abs. air; 4.21 ion. ch.		Natural source, parent of UX <sub>1</sub> <sup>234</sup>
U <sup>239</sup>	A		$\beta^-$ , $\gamma$ , $e^-$	23.5 min; 23 min; 23.2 min; 23.54 min	1.20 abs. Al; 1.2 abs. Al; 1.12, 2.06 (weak) spect.	0.076, > 0.3 (weak) abs. Pb; 0.073, 0.92 spect. conv., abs. Pb	U-n- $\gamma$ , parent of Np <sup>239</sup> U-d-p
93 Np <sup>231</sup>	A		$\alpha$ , K	53 min	6.2( $\alpha$ ) ion. ch.		U-d-9n, parent of Pa <sup>237</sup> U <sup>234</sup> -d-6n U <sup>234</sup> -d-4n
Np <sup>234</sup>	B		K, $\gamma$	4.40 days; 4.4 days		1.9 abs. Pb	Pu <sup>234</sup> K-de- cay(?) U <sup>234</sup> -d-n U <sup>234</sup> -d-3n Pa- $\alpha$ -n U <sup>234</sup> - $\alpha$ -p2n U <sup>234</sup> - $\alpha$ -p4n U <sup>234</sup> -p-2n
Np <sup>235</sup>	B		K; $\alpha$ ( $\sim 0.1$ %)	435 days; 400 days	5.06( $\alpha$ ) ion.ch.	No $\gamma$ (?)	U <sup>234</sup> -d-2n U <sup>234</sup> - $\alpha$ -pn U <sup>234</sup> - $\alpha$ -p3n Parent of Pu <sup>236</sup> U <sup>234</sup> -d-n U-d-4n U <sup>234</sup> - $\alpha$ -p U <sup>234</sup> - $\alpha$ -p2n Np-d-t Np- $\alpha$ - $\alpha$ n U <sup>237</sup>
Np <sup>236</sup>	A		$\beta^-$ , $\gamma$	22 hr	0.5 abs. Be		$\beta^-$ -decay, parent of Pa <sup>233</sup>
Np <sup>237</sup>	A		$\alpha$	$2.20 \times 10^4$ yr; $3 \times 10^4$ yr	4.77 ion.ch.; 4.75 abs. air; 4.73 abs. Al;		

Isotope $Z$ $A$	Class	Per cent abundance	Type of radiation	Half-life	Energy of radiation, Mev		Produced by
					Particles	$\gamma$ -rays	
93 Np <sup>237</sup>							
Np <sup>238</sup>	A		$\beta^-$ , $\gamma$ , $e^-$	2.10; 2.0 days	4.72 abs. mica 0.22, 1.39 abs. Al; 1.0 abs. Al	1.2, 0.075 abs. Pb, abs. of $e^-$ ; 1.1 abs. Pb	Parent of Pu <sup>238</sup> U <sup>238-d-2n</sup> U-d-2n Am <sup>242</sup> $\alpha$ -decay U- $\alpha$ -p3n U- $\alpha$ -p Np-n- $\gamma$ Np-d-p U <sup>239</sup> $\beta^-$ -decay, parent of Pu <sup>239</sup> U-d-n U- $\alpha$ -p2n
Np <sup>239</sup>	A		$\beta^-$ , $\gamma$ , $e^-$	2.33 days; 2.3 days; 2.35 days	0.68, 0.33, 0.090 abs.; 0.47 abs.; 0.14, 0.40, 0.63 abs. Al; 0.78 abs. Al; 0.288, 0.403, 0.673, 1.179 spect.	0.057, 0.061, 0.067, 0.206, 0.227, 0.275 spect. conv.; 0.2094, 0.2280, 0.2774, numerous softer $\gamma$ 's spect. conv.; 0.22, 0.27 spect. conv., spect.	
94 Pu <sup>232</sup>	B		$\alpha$	22 min	6.6 ion.ch.		U <sup>238</sup> - $\alpha$ -7n, parent of U <sup>238</sup>
Pu <sup>234</sup>	A		$\alpha$ ; K	8 hr; 8.5 hr	6.2 ion.ch.; 6.0 ion.ch.		U <sup>232</sup> - $\alpha$ -3n, parent of U <sup>230</sup> and Np <sup>234</sup> (?) Np <sup>236</sup> $\beta^-$ -decay Cm <sup>240</sup> $\alpha$ -decay, parent of U <sup>232</sup> U <sup>238</sup> - $\alpha$ -3n U <sup>232</sup> - $\alpha$ -12 U- $\alpha$ -6n Np- $\alpha$ -p4n Np-d-3n U <sup>238</sup> - $\alpha$ -2n U- $\alpha$ -5n Np-d-2n
Pu <sup>236</sup>	A		$\alpha$	2.7 yr	5.75 ion.ch.; 5.7 ion.ch.		
Pu <sup>237</sup>	B		K	40 days		No $\gamma$	



Isotope Z     A	Class	Per cent abundance	Type of radiation	Half-life	Energy of radiation, Mev		Produced by
					Particles	$\gamma$ -rays	
94 Pu <sup>238</sup>	A		$\alpha$	92 yr; 89 yr; 40 yr	5.51 abs. air; 5.5 abs. air, Al; 5.4 abs. Al; 5.493 ion.ch.		Np <sup>238</sup> $\beta^-$ -decay Cm <sup>242</sup> $\alpha$ -decay Np-d-n U- $\alpha$ -4n U <sup>238</sup> - $\alpha$ -n
Pu <sup>239</sup>	A		$\alpha, \gamma, e^-$	$2.411 \times 10^4$ yr calor.; $2.44 \times 10^4$ yr	5.15 abs. air; 5.1 abs. air; 5.16 cl.ch.; 5.140 ion. ch.; 5.159 ion.ch.	0.42, 0.2 (weak) abs. Pb; 0.05, 0.3 (weak) abs. Pb, Al	Np <sup>239</sup> $\beta^-$ -decay Natural source U- $\alpha$ -3n
Pu <sup>240</sup>	A		$\alpha$	$\sim 6000$ yr yield	5.1 ion.ch.		U- $\alpha$ -2n
Pu <sup>241</sup>	A m.s.		$\beta^-; \alpha$ ( $\sim .002\%$ )	$\sim 10$ yr yield	0.01-0.02 ( $\beta^-$ ) abs. hydro- carbon; 5.0( $\alpha$ ) calc.		U- $\alpha$ -n, parent of Am <sup>241</sup> and U <sup>237</sup>
95 Am <sup>238</sup>	D		K(?)	1.5 hr			Pu-d-3n
Am <sup>239</sup>	B		K( $\sim 100\%$ ), $e^-, \gamma$ ; $\alpha$ ( $\sim 0.1\%$ ),	12 hr	5.77( $\alpha$ ) ion.ch.	0.285 abs. Pb, abs. of $e^-$	Pu-d-2n Pu-p-n Np- $\alpha$ -2n
Am <sup>240</sup>	B		K, $\gamma, e^-$	50 hr; 53 hr		1.3 abs. Pb, abs. of $e^-$	Pu-d-n Np- $\alpha$ -n
Am <sup>241</sup>	A		$\alpha, \gamma$	490 yr; 510 yr	5.48 ion.ch.; 5.45 ion. ch.	0.062 abs. Pb	Pu <sup>241</sup> $\beta^-$ -decay
Am <sup>242m</sup>	A		$\beta^-$	16 hr; 17 hr	0.8 abs. Al		Am-n- $\gamma$ , parent of Cm <sup>242</sup>
Am <sup>242</sup>	A		$\alpha$ ( $\sim 0.2\%$ ), $\beta^-$	$\sim 400$ yr	$\sim 0.5$ ( $\beta^-$ ) abs. Al; 5.2( $\alpha$ ) calc.		Am-n- $\gamma$ , parent of Cm <sup>242</sup> and Np <sup>238</sup>
96 Cm <sup>238</sup>	B		$\alpha$	$\sim 2.5$ hr	6.50 ion.ch.		Pu- $\alpha$ -5n
Cm <sup>240</sup>	A		$\alpha$	26.8 days	6.26 ion.ch.; 6.3 ion.ch.		Pu- $\alpha$ -3n, parent of Pu <sup>238</sup>
Cm <sup>241</sup>	E		K	55 days			Pu- $\alpha$ -2n
Cm <sup>242</sup>	A		$\alpha$	150 days	6.08 ion.ch.; 6.1 ion.ch.		Pu- $\alpha$ -n Am <sup>242</sup> and Am <sup>242m</sup> $\beta^-$ -decay, parent of Pu <sup>238</sup>



## PART II

# Methods and Instruments



## CHAPTER 8

### INDIRECT METHODS FOR MEASURING DEUTERIUM

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**8.1. Introduction.** The stable isotopes offer the advantage of avoiding the time limits inherent in many radioactive-isotope experiments. Their abundance can be determined directly with the mass spectrometer (Chap. 9) or indirectly from measurements of those physical properties which vary with isotopic ratio. In general, the detection of small differences in these properties is difficult, and in those cases where there is relatively little difference in the masses of the isotopes involved, even very large changes, say 100 per cent, in isotopic composition cause undetectably small changes in these properties. This, with the added complication that traces of impurities may exert comparatively large effects, has so far served to render these methods useless for all but two isotopes, deuterium and the heavy isotope of oxygen,  $O^{18}$ .

Among the methods based on density differences which have been developed for analysis of deuterium in water are the use of the pycnometer [1,2], the free float [3-10], the falling drop [11-14], and the diffusion gradient [15,16,17]. Methods based on changes in the refractive index [18,19] and in the vapor pressure [74] of water have also been developed for deuterium analysis. The vapor-pressure method has a lower precision than the other methods, but does afford a rapid means for an approximate  $D_2O$  analysis on a minute amount of material.

A wide assortment of methods based on changes in other physical properties of water or of hydrogen gas have been used with varying degrees of success. Among these are spectrum analysis [20], thermal conductivity [20-23], the deuterium-lithium nuclear reaction [20,24], electrode potentials [25], viscosity [26], freezing point [27], infrared absorption [28], and gas-density balance [60,61]. Some of these obviously are not suitable for routine applications.

Density methods have also been used to determine the abundance of  $O^{18}$  in water [29].

**8.2. Physical Properties of Deuterium Oxide and Water.** In this section data available prior to 1941 are summarized. Because of the importance of heavy water in atomic power very little of such information has appeared in the open literature since that date.

Some of the physical properties of deuterium oxide and of natural water are compared in Table 28, abstracted from a more extensive tabulation in the

review by Urey and Teal [20], in which references to the original data may be found. The values for density and the temperature of maximum density have been changed in accordance with more recent data [30-32].

Water has its maximum density at 3.98°C. The volume of 0.001 liter of water at this temperature is, by definition, 1 milliliter. Density measurements are commonly expressed as grams of weight per milliliter and therefore are called "density (or specific gravity) relative to water at 4°C." The notation  $d_4^t$  signifies that the weight of a given volume at the temperature indicated by the superscript is referred to that of water at 3.98°C, and therefore also indicates the weight per milliliter directly. These values are slightly different from those of absolute density, which conform to the cgs system and express density as grams of mass per cubic centimeter. Similarly, the notation  $d_{25}^{25}$  indicates that the weight of a certain volume of the sample at 25°C is referred to that of the same volume of water at 25°C.

TABLE 28. SOME PROPERTIES OF H<sub>2</sub>O AND D<sub>2</sub>O  
Abstracted from Urey and Teal [1935], [20].

Property	H <sub>2</sub> O	D <sub>2</sub> O
Density, $d_{25}^{25}$ .....	1.0000	1.10764*
Temperature of maximum density.....	3.98	11.23 ± 0.02*
Molar volume at temperature of maximum density.	18.015 cc	18.140 cc
Surface tension.....	72.75 dynes/cm	67.8 dynes/cm
Viscosity in millipoises:		
10°C.....	13.10	16.85
20°C.....	10.09	12.60
30°C.....	8.00	9.72
Refractive index, 20°C Na D line.....	1.33300	1.32828
Melting point.....	0.0	3.802
Boiling point.....	100.0	101.42

\* More accurate values than in original table [30-32].

The molar volume is, of course, the volume of a gram-molecular weight. If the molar volume of deuterium oxide and water were equal, the  $d_{25}^{25}$  of deuterium oxide would be 1.1116 instead of 1.10764.

Table 29 presents the combined data of Longworth [33], Swift [31,34], and Tronstad *et al.* [30], with Longworth's values for mole fraction recalculated to conform to a pure deuterium oxide  $d_{25}^{25}$  of 1.10764. The values of  $d_4^{25}$  are the experimental values obtained by Swift and by Longworth. The corresponding  $d_{25}^{25}$ 's were calculated from these. The final column gives the density of each dilution at its temperature of maximum density.

These data may be used to show that D<sub>2</sub>O-H<sub>2</sub>O mixtures form almost ideal solutions, *i.e.*, there is a negligible volume change upon mixing (the density of an ideal solution is not necessarily a linear function of the mole

fraction, but the molecular volume is). Other evidence shows the solutions to be ideal in the very low (tap-water) deuterium oxide concentration range, [35] quoted in [31]. At 25°C the equation [31]

$$N_{D_2O} = \frac{9.257\Delta S}{1 - 0.033\Delta S}$$

where  $N_{D_2O}$  = mole fraction of deuterium oxide and  $\Delta S = d_{25}^{25} - 1$  relative to deuterium-free water, may be used to calculate the mole fraction of deuterium in a sample from the density of the sample. This equation in the form

$$d = 1 + \frac{N_{D_2O}}{9.257 + 0.033N_{D_2O}}$$

was used to calculate Table 30.

The abundance of deuterium oxide in nature has been the object of extensive research [10,22,36]. Long before the discovery of isotopes a variation of 8 parts in  $10^7$  was found in the densities of various samples of water that were expected to be identical. The average ratio of  $D_2$  to  $H_2$  is about 1:6,500 [51,52]. The issue is confused by variations in the abundance of  $O^{18}$  which produces similar variations in density [29,38-43]. The accepted ratio for the relative abundance of the oxygen isotopes in 1941 was

$$O^{16}:O^{18}:O^{17}::(506 \pm 10):1:(0.204 \pm 0.008) [54]$$

Commercial oxygen obtained from liquid air was found to have an atomic weight  $2.2 \times 10^{-6}$  unit heavier than atmospheric oxygen. That obtained by electrolysis is lighter, the fractionating factor being 1.008 [40]. The atomic weight of atmospheric oxygen is  $11.9 \times 10^{-6}$  unit greater than that in water, and the average density for water made from air is  $6.4 \times 10^{-6}$  gm per ml heavier than ordinary water [41]. Lewis and Luten [18] show that from determination of both the refractive index and the density of a water sample the concentrations of both  $O^{18}$  and deuterium may be found by solving two simultaneous equations relating the effects.

Tables of the density of water to seven decimal places at temperatures from 0 to 40°C by 0.1°C intervals are available, but Dorsey [37] questions the accuracy of such tables because variations in deuterium oxide content exceed the precision indicated.

At room temperature the density of water decreases by about 0.03 per cent per degree centigrade rise in temperature. Table 31 lists the densities  $d_4^t$  of natural water and of deuterium oxide. The values up to 42°C in the column for water were obtained by rounding off to six places the values from Dorsey [37]. Comparison with the last two columns [30,44] shows that deuterium oxide does not expand with temperature at quite the same rate as does water.

TABLE 29. DENSITIES OF MIXTURES OF  $\text{H}_2\text{O}$  AND  $\text{D}_2\text{O}$ 

Combined experimental data of Longworth [33], Swift [31,34], and Tronstad *et al.* [30].  
Using  $\text{D}_2\text{O}$ ,  $d_{25}^{25} = 1.10764$  (referred to natural water).

Mole fraction % of $\text{D}_2\text{O}$	$d_4^{25}$	$d_{25}^{25}$	Temp at max density	$d_4^t$
Pure $\text{D}_2\text{O}$				
100.00	1.10439	1.10764	$11.23 \pm 0.02$	1.10596
99.85	1.10422	1.10746	11.1	1.10577
99.59	.....	.....	11.1	1.10522
99.42	1.10376	1.10700		
93.53	1.09746	1.10068	10.8	1.109909
82.55	1.08570	1.08890		
77.80	1.08083	1.08401	9.7	1.08264
61.16	1.06279	1.06591		
60.51	1.06208	1.06520	8.7	1.06411
40.34	1.04044	1.04349		
37.84	1.03777	1.04082	7.0	1.04019
20.24	1.01884	1.02183		
18.66	1.01715	1.02014	5.6	1.01978
Natural water				
00.017	0.99707	1.00000	3.85	1.00000
$\text{H}_2\text{O}$				
00.000	0.99705	0.99998		

TABLE 30. DENSITIES OF MIXTURES OF  $\text{D}_2\text{O}$  AND  $\text{H}_2\text{O}$  AT  $25^\circ\text{C}$   
Calculated [31] from the formula

$$d_{25}^{25} = 1 + \frac{N}{9.257 + 0.033N}$$

Mole fraction % of $\text{D}_2\text{O}$	$d_{25}^{25}$	Mole fraction % of $\text{D}_2\text{O}$	$d_{25}^{25}$
100.00	1.1076	45.00	1.0485
95.00	1.1023	40.00	1.0431
90.00	1.0969	35.00	1.0378
85.00	1.0915	30.00	1.0324
80.00	1.0862	25.00	1.0270
75.00	1.0808	20.00	1.0216
70.00	1.0754	15.00	1.0162
65.00	1.0701	10.00	1.0108
60.00	1.0647	5.00	1.0054
55.00	1.0593	0.00	1.0000
50.00	1.0539		



The difference in density goes through a maximum at about 40°C. The precision of temperature control required will be mentioned in the discussions of the individual methods. The techniques for thermoregulation are discussed under the falling-drop method.

TABLE 31. DENSITIES OF NATURAL WATER AND D<sub>2</sub>O

<i>t</i>	$d_4^{25} \text{H}_2\text{O}$ [37]	$d_4^{25} \text{D}_2\text{O}$ [30]	$d_4^{25} \text{D}_2\text{O}$ [44]
5	0.999992	.....	1.10549
10	0.999728	1.105948	1.10588
12	0.999526	1.105956	
14	0.999273	1.105891	
15	0.999129	.....	1.10577
16	0.998972	1.105755	
18	0.998625	1.105558	
20	0.998234	1.105300	1.10527
22	0.997800	1.104981	
24	0.997327	1.104608	
25	0.997075	1.104400	1.10440
26	0.996814	1.104180	
27	0.996544	1.103947	
30	0.995678	.....	1.10321
35	0.994064	.....	1.10172
38	0.992997	.....	1.10068
39	0.992626	.....	1.10032
40	0.992247	.....	1.09995
41	0.991861	.....	1.09956
42	0.991468	.....	1.09917
43	0.9911	.....	1.09876
45	0.9902	.....	1.09792
50	0.9880	.....	1.09572

A pressure increase of 1 atm increases the density of water by about 0.005 per cent. Organic liquids are affected by about the same order of magnitude. It is apparent, therefore, that the ordinary daily fluctuations of pressure (about  $20/60$  atm) are negligible in almost all density determinations.

Dissolved air also decreases the density of water. Various workers do not agree on the extent of this effect, but at 20°C the difference in density between air-free water and water that has been in contact with air for 3 or 4 days is about  $2 \times 10^{-6}$  to  $2 \times 10^{-7}$  [37].

**8.3. Preparation and Purification of Water Samples for Analysis.** Organic compounds are analyzed for deuterium content by burning them to form water, collecting and purifying the water formed, and determining the deuterium oxide content of this water [13,14]. Combustion is carried out in a quartz or Vycor tube packed with cupric oxide and heated to 750°C. Oxygen manufactured from liquid air is bubbled through sulfuric acid and

passed through a trap immersed in solid carbon dioxide before entering the combustion tube. The water formed is collected by condensation in another trap surrounded by solid carbon dioxide.

Purification and analysis of water should be conducted in a room free from vapors of organic solvents. A system for purification is illustrated by the distillation train shown in Fig. 54. It consists of a series of traps made from 10-mm glass tubing. The sample to be purified is placed in the first trap *A*. The second trap contains a few milligrams of dry chromic acid and the third a few milligrams of potassium hydroxide, or better, calcium oxide, which avoids replaceable hydrogen, and potassium permanganate.

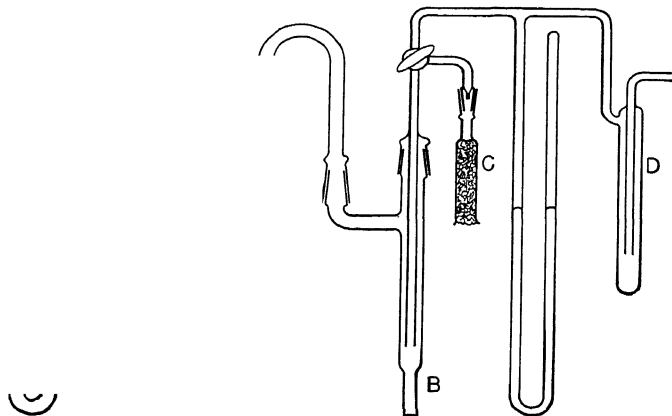


FIG. 54. Vacuum distillation train.

Vacuum distillation is accomplished by chilling the second trap with solid carbon dioxide and evacuating the system. Distillation is accelerated by heating the first trap with water in a beaker or with a couple of 3,500-ohm resistors connected to the 110-volt a-c supply. Vapors from the pump are prevented from entering the system by interposing another carbon dioxide-chilled trap (*D*, Fig. 54). After the water has been collected in the second trap, air is admitted through the drying tube *C* containing magnesium perchlorate, the speed of flow being reduced by a capillary. The first trap is removed and replaced by a stopper, the water in the second trap is melted and gently boiled, then the third trap is chilled and the water distilled into it. Upon collection in the final trap *B*, the water is ready for analysis.

A simpler method for small samples of water involves placing the sample in the bottom of a small petri dish or ampule and allowing it to condense and freeze on the inside of the cover, which is chilled with dry ice. The bottom is then removed, the cover inverted and put in its place, a clean cover put

in the position of the first one, and the process repeated for several redistillations [73].

It is imperative that all glass with which the water comes in contact be scrupulously cleaned. The recommended procedure is to wash all the articles concerned with hot solutions of trisodium phosphate, rinse with distilled water, treat with boiling concentrated nitric acid, rinse about ten times with freshly distilled water, and finally dry the articles in an electric oven free from organic materials.

**8.4. Refractive Index.** For the sodium D line, the refractive index  $n$  of a mixture of deuterium oxide and water at 20°C lies between that of pure deuterium oxide, 1.32828, and that of ordinary water, 1.33300, and is a linear function of the mole fraction. In order to use this property in the estimation of the deuterium content of a sample, one must determine its value very exactly. The simpler types of refractometers are not suitable for such determinations, but the interferometer can give adequate precision.

As its name implies, the interferometer is designed to utilize the interference of light waves. The difference between the refractive indices of two samples is found from the shift in the position of interference bands. Instruments are available commercially with which an accuracy of  $\pm 1 \times 10^{-7}$  can be obtained, and with certain modifications, the accuracy can be extended to  $\pm 1 \times 10^{-8}$  [46]. Because it is a differential instrument, the temperature requirements are not nearly so exacting as in other instruments; no special temperature control is required for an accuracy of  $\pm 1 \times 10^{-7}$ . However, errors due to impurities in the sample may be great. There are a number of indeterminate errors so that the measurements of  $\Delta n$  are not absolute and calibration is necessary.

The Columbia University group has used a Zeiss interferometer calibrated to read directly in per cent of deuterium [13,14,45,47]. They find that the deuterium content of 0.4 cc of water can easily be determined to an accuracy of 0.02 atom per cent.

The calibration of the instrument is accomplished by making determinations on samples of known composition. This is a tedious task, but when done provides a rapid and satisfactory method of analysis.

**8.5. Pycnometer.** The pycnometer method usually serves as the primary standard for other methods of deuterium analysis. Because it is tedious—one determination may take an entire day—and because it requires large samples (5 to 50 ml), it is not suited to routine determinations.

The methods commonly used for accuracies of 1 part in  $10^6$  are usually modifications of the differential method described in detail by Washburn and Smith [1,2]. In these methods twin silica pycnometers of the type illustrated in Fig. 55 are employed, having a bulb volume of 50 cc and a capillary diameter of 0.1 cm. The capillary stem of each has a thin reference mark  $A$ ,

and the volume per unit length of the capillary is determined by calibration with mercury.

To find the difference in density between a given sample and normal water the first pycnometer is filled with water and the second with the sample to suitable heights in the capillaries. Filling is conveniently done with a fine silver capillary and the aid of a low vacuum line, as illustrated in Fig. 56. The pycnometers are then placed side by side, with their ground-glass stoppers loosely in place, in a thermostat held constant to  $\pm 0.01^\circ\text{C}$ . After thermal equilibrium is attained, the height of the meniscus above the reference mark is read with a cathetometer to 0.001 cm. The pycnometers are next taken out of the bath, wiped dry, placed on opposite pans of a microbalance with

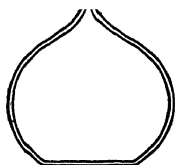


FIG. 55. Pycnometer.

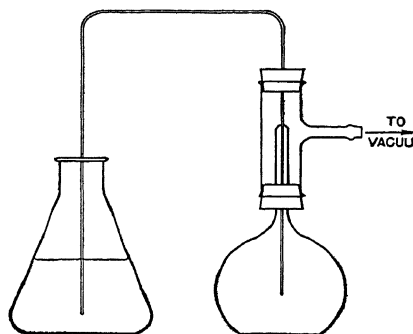


FIG. 56. Device for filling pycnometer.

their stoppers tightened in place, and the difference in their weights is determined to 0.03 mg. After the weighing, they are emptied and the procedure is repeated with the sample in the first pycnometer and normal water in the second one and with the pycnometers placed on the same sides of the balance as before. The weights, volumes, and densities involved are simply related by two equations which are solved for the difference in density.

This use of twin pycnometers avoids, by balancing them out, many of the sources of error and corresponding corrections necessary in other techniques, such as the effect of humidity, buoyancy of the pycnometer, effect of barometric and hydrostatic pressure on the capillary, and the effect of dissolved air. The temperature of the thermostat need not be accurately known, but it should be constant and uniform while the pycnometers are in the bath. The small correction for the deviation from a linear relation between the mole fraction and the density is made [31] by using the following equation:

$$N_{D_2O} = \frac{9.257\Delta S}{1 - 0.033\Delta S}$$

where  $N_{D_2O}$  = mole fraction of  $D_2O$

$$\Delta S = d_{25}^{25} - 1 \text{ relative to D free water}$$

**8.6. Free Float.** Several methods of varying the upward buoyant force exerted on a body immersed in a liquid have been sufficiently developed to serve as extremely sensitive methods for measuring small differences in density. These differ among themselves chiefly in the manner in which the effective density of a glass bulb is varied with respect to the solution in which it is immersed.

Most successful with regard to the degree of accuracy attained is the method of Lamb and Lee [3,46]. A small piece of soft iron is sealed in the bulb and a copper solenoid wound around a beaker containing the sample. The passage of current through the solenoid produces a magnetic force that causes the bulb to sink. At the bottom, the bulb makes contact with a platinum electrode. The current is then adjusted just enough to break the contact, and a reading is taken on a milliammeter. The greater the density of the liquid, the greater is the current required to balance the upward buoyant force. An accuracy of  $1 \times 10^{-7}$  in  $\Delta d$  was obtained by the original method and the extremely high accuracy of  $\pm 2 \times 10^{-8}$  in  $\Delta d$  with subsequent improvements.

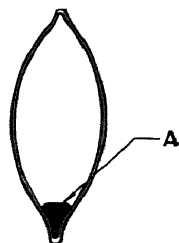


FIG. 57. Float used in free-float methods.

Another method [4-6] uses a float (Fig. 57) weighted with mercury, *A*, and depends upon varying the temperature and determining that critical temperature at which the float will neither rise nor sink. Precise thermometry is basic to the method inasmuch as a change of  $0.001^\circ C$  will cause the change from rising to sinking. A change of  $1 \times 10^{-6}$  in density changes the equilibrium about  $0.006^\circ$ . A plot of equilibrium temperature against density gives almost but not quite a straight line.

A somewhat more convenient method employs the Cartesian diver principle in which the temperature is held constant but the hydrostatic pressure on the system is varied [7-10,13]. A float similar to the one in the previous method (Fig. 57) is sealed in a tube connected to a pump and manometer system. The sample is distilled in vacuo into the tube containing the float. It is practical to adjust the density of the float to correspond to that of water at  $4.58^\circ C$ , at which temperature the thermal expansion of water is equal to that of pyrex glass and small temperature fluctuations produce no appreciable change in the relative densities. The float is mounted in a well-stirred thermostat maintained at  $4.58^\circ C$  by the flow of water through the thermostat. The flow of water is regulated by a valve operated by a thyatron

control system. If the float is more compressible than water, a rise in pressure increases its relative density and causes it to sink. The motion of the float is observed by a wide-field telescope with a scale in the ocular. The equilibrium pressure is deduced from the rate of drift at pressures a little low and a little high. With the pressure set at this value, the float will remain stationary for as long as 15 min. In one of the setups of this general type [13], a pressure change of 1 cm of mercury corresponded to a density change of  $0.376 \times 10^{-6}$ . The method has been used for measurement of deuterium concentrations below 0.03 per cent, and a difference of 0.0002 atom per cent of deuterium can be determined, corresponding to a change in density of  $2 \times 10^{-7}$  [48].

Like the pycnometer method, these buoyancy methods have the disadvantage of being tedious and not readily adaptable to rapid determinations.

**8.7. Falling Drop.** In the falling-drop method the density of a sample of water is determined by measuring the rate at which a drop of the sample falls through an immiscible liquid of slightly lower density. This liquid, the reference medium, is contained in a tube mounted vertically in a thermostatically controlled water bath. The drops are formed with a mechanical micropipette, and the rate of fall is determined by timing the interval that elapses while the drop falls the distance between two fiducial marks on the tube.

For low velocities, the rate of fall is determined by a number of factors as related in Stokes' law

$$6\pi\eta av = \frac{4}{3}\pi a^3(d - d_o)g$$

where  $\eta$  = viscosity of medium

$a$  = radius of drop

$v$  = velocity of drop

$d$  = density of drop

$d_o$  = density of medium

$g$  = acceleration due to gravity

In order to obtain accurate and reproducible determinations of the density from the rate of fall, it is essential to choose a medium, drop size, and operating temperature that will give falling times short enough for convenience but with sufficient spread to give the required sensitivity over the range of densities that will be encountered and to keep these factors constant.

The originators of this method used a mixture of bromobenzene and xylene as the medium [11,12]. By varying the proportions of these substances one can obtain mixtures having a wide range of densities. The differential vaporization that occurs, however, causes the composition of the mixture to change from day to day. Keston *et al.* [14] introduced the use of *o*-fluorotoluene, which has a density convenient for measuring deuterium oxide con-

centrations of 0 to 3 per cent at 26.8°C and which avoids the differential vaporization of a binary system. *m*-Fluorotoluene may be used for the same concentration range at 19.3°C. These toluenes have the further advantage of lower viscosity and therefore greater sensitivity. They may be obtained from the Eastman Kodak Company or synthesized by a procedure analogous to that for fluorobenzene [49].

For higher concentrations of deuterium oxide, *o*-fluorotoluene is unsatisfactory because the precision of the method falls off rapidly as the difference between the density of the water and the reference liquid increases. For samples containing 10 to 40 per cent deuterium oxide, solutions of phenanthrene in  $\alpha$ -methylnaphthalene have been shown to be superior reference liquids [50]. Neither of these components is particularly volatile. The exact viscosity of *o*-fluorotoluene has not been published, but it is estimated that that of  $\alpha$ -methylnaphthalene is about three times as large; so for the same density difference, the drops fall more slowly in the latter medium.

The dimensions of the tube containing the medium are not critical except that the inside diameter should be greater than three times the diameter of the drop to avoid wall effects. The fiducial marks should be as far apart as possible (about 20 cm) to provide maximum sensitivity and the best average rate of fall. Another 20 cm should be allowed above the top mark to ensure thermal equilibrium between the drop and the medium by the time the drop reaches the first mark, and about 10 cm should be allowed below the lower mark to avoid end effects.

The size of the drop is controlled through the use of a mechanical micropipette. The size is not critical and may be 5 to 45 mm<sup>3</sup> [54], but it is imperative that it be uniform in any set of determinations, including the associated calibration. It is desirable to use as small a drop as possible to minimize the quantity of water needed for an analysis, but the sensitivity of the method increases with the square of the radius [54]. Variations in drop size introduce a percentage error in velocity which is two-thirds the percentage error in volume. For falling velocities of the order of 1 cm per sec, the variation in drop size must be less than 0.1 per cent.

The designs for several satisfactory micropipettes have been published [14,55-58]. The accompanying drawing (Fig. 58) illustrates the one used by the authors [59]. It is somewhat less bulky than other published instruments but provides the required precision. The barrel *K* is made from a 1/4-cc glass tuberculin syringe and is joined to the capillary with cobalt glass. Mercury is displaced by the advancement of a piece of 1/16-in. diameter drill rod driven by a micrometer head. A mechanical stop *A* assures uniformity of the angle through which the micrometer is turned. With the sample in the capillary, the tip is lowered about 1 cm below the surface of the medium and a drop formed. The drop is released by raising the pipette slowly and

allowing the surface tension of the medium to pull the drop from the capillary tip.

The relative density of water and the medium vary so greatly with temperature that the temperature must be kept constant to  $\pm 0.001^\circ\text{C}$ . This is accomplished by mounting the tube in a well-stirred water bath of about 100 liters capacity and carefully controlling the bath temperature. Some

form of automatic temperature regulation is required. Baths with the required control are available commercially. Many designs for thermostats have been published [62-68]. If the bath temperature is to be above room temperature, a temperature-sensitive element such as a volume of mercury with a large surface-volume ratio or a bimetallic strip may be used to control the current of an immersion heater through a relay or thyatron circuit. In a thermostat that uses a relay, the discontinuous input is the fundamental obstacle to the attainment of perfect constancy of temperature. Amplifier devices may be used to give a continuous but variable input, but these also have a finite sensitivity. The Guoy principle, applying a vertical oscillating motion to the contacting wire of an expansion thermoregulator, provides an intermediate type of control between simple on-and-off control and continuous control and has been used to hold the temperature of a 100-liter water bath constant within  $\pm 0.0002^\circ\text{C}$  for several hours [66]. There is some advantage in having the

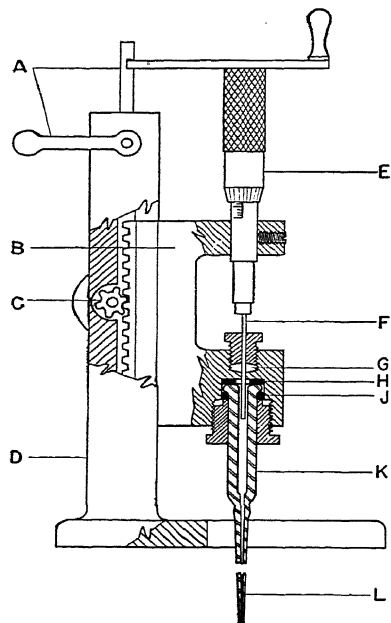


FIG. 58. Micropipette. *A*, stop; *B*, bracket; *C*, pinion; *D*, stand; *E*, micrometer; *F*, plunger ( $\frac{1}{16}$  in. diameter drill rod); *G*, packing; *H*, rubber washer; *J*, lead washer; *K*, barrel; *L*, capillary.

bath temperature below room temperature [56]. This may be effected with the same kind of thermoregulators, but by controlling the flow of cold water through the bath instead of controlling a heater. Many difficulties are avoided by having the apparatus in a constant-temperature room and operating near room temperature. The multiple-enclosure principle of Tian may also be used to decrease the effects of ambient variations [68,69].

Under the conditions used in this method, the rate of fall does not follow Stokes' law perfectly, and it is necessary to establish a calibration curve by



plotting the densities of several mixtures of known density against the reciprocals of their falling times. The deviation from Stokes' law may be calculated from the differences between the reciprocals of the times of fall of the known mixtures and that of pure water [54].

Depending upon the range of concentrations being determined, this method gives a precision in density of about 1 to 4 parts in  $10^6$ , corresponding to 0.001 to 0.004 per cent of deuterium. The errors introduced in the combustion and purification processes are larger and may raise the maximum over-all error to 0.02 per cent of deuterium.

**8.8. Diffusion Gradient.** This method is similar to the falling-drop method in that the density of a drop of the liquid to be tested is determined by introducing the drop into another liquid with which it is immiscible. It differs chiefly in that the reference liquid is so prepared that it presents a linear gradient of density and the drop falls only to an equilibrium position at which the specific gravity of the surrounding medium is equal to its own. The density of the drop is found by comparing the equilibrium position of the unknown sample with the positions of other drops of known density. This method offers the advantage of avoiding the errors due to convection currents inherent in the falling-drop method [15-17,70].

Figure 59 shows the type of tube used in this method [15]. The tube is mounted in a vertical position to the depth indicated by *A* in a thermostated water bath. A gradient suitable for determination of the deuterium in deuterium oxide-water mixtures containing between 0 and 10 per cent deuterium oxide may be prepared from two bromobenzene-kerosene mixtures, the lighter one having a density of about 0.99 and the heavier a density of about 1.02 [70].

The tube is placed in the thermostat (regulation to  $\pm 0.01$  is sufficient), and the heavier mixture is poured in up to the middle of the tube connecting the two bulbs. Next the lighter mixture is brought to the bath temperature and added cautiously through a funnel with a filter up to the level shown at *B* in Fig. 59. In order to obtain a uniform distribution of the two mixtures in

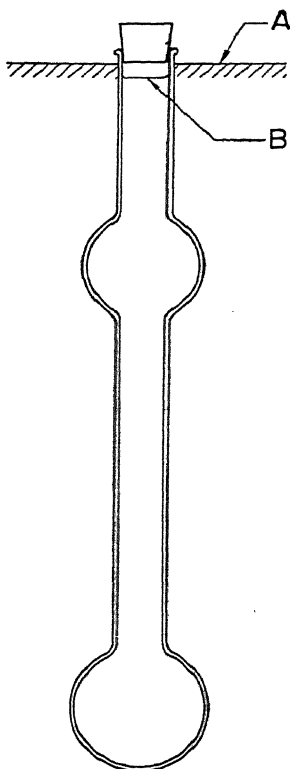


FIG. 59. Tube for diffusion gradient.

the connecting tube, a stirrer is lowered to the center of the tube and slowly pulled up and down in the tube for about ten cycles. After standing about 48 hr, the gradient is sufficiently linear to be used in carrying out measurements. The early papers on this method [15] recommended saturating the kerosene-bromobenzene mixture with water at a suitable vapor pressure, but in deuterium measurements this is undesirable because of exchange. Although it is true that if the gradient is used exclusively for deuterium analysis the various levels of the gradient do tend to become saturated with the proper deuterium oxide-water ratios, exchange may still occur while the drop is settling, and it is best to keep the gradient unsaturated.

The tube is calibrated by determining the equilibrium positions of drops of known density. In use, several drops covering the density range in which the experimental drops are expected to lie are introduced in the left side of the tube and an experimental drop on the right side. After 15 min the relative positions are determined with the aid of a cathetometer. As in the falling-drop method, the size of the drops is not critical within reasonable limits (about 0.1 to 10 mm<sup>3</sup>), but those used must be uniform. The pipette can be somewhat simpler than the type needed for the falling drop, the micro-constriction type being satisfactory [71].

As described above, this method gives a precision of only  $\pm 0.1$  per cent of deuterium, but by simultaneously increasing the drop size and decreasing the range of the gradient (using mixtures of densities 0.995 and 1.005, for example), a precision of 5 parts in 10<sup>6</sup> can be achieved [72].

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## CHAPTER 9

### MASS SPECTROGRAPHS

**9.1. Aston Mass Spectrograph.** An arrangement of electric and magnetic fields devised and perfected by Aston [1,2] was the first employed for extensive and accurate measurements of isotope masses and relative abundances. A highly collimated ion beam emerging from the slits  $S_1$  and  $S_2$ , as shown in

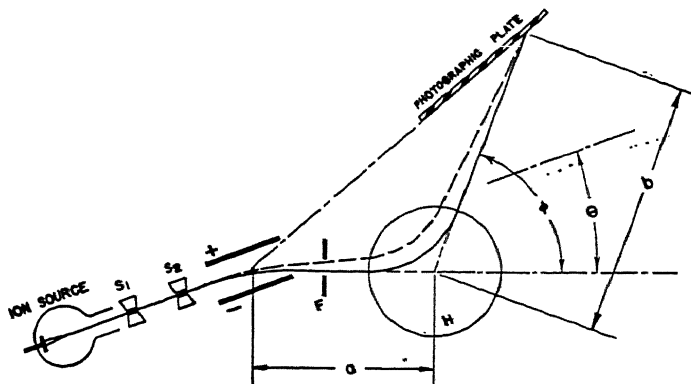


FIG. 60. Aston mass spectrograph.

Fig. 60, passes through a strong uniform electric field between two parallel electrodes which deflects the ions by a small angle

$$\theta = \frac{lV}{v^2} \frac{e}{m}$$

where  $l$  = length of path between plates

$V$  = deflecting voltage on plates

$v$  = initial ion velocity

$e$  = ion charge

$m$  = ion mass

The defining vane  $F$  then permits only ions within a small prescribed range of velocities to enter the uniform magnetic field  $H$ . The angular deflection in the magnetic field is given by the expression

$$\phi = LH \left( \frac{\theta e}{lV m} \right)^{1/2}$$

where  $L$  = length of path in magnetic field

Separation of ions with different values of  $m/e$ , as well as focusing of identical values, takes place only in the magnetic field. The electrostatic field serves only as a velocity analyzer to focus the inhomogeneous ion beam into an energy spectrum from which only a small portion is selected by the defining vane and allowed to enter the magnetic field.

For small deflections, the focal points for different values of  $m/e$  lie along the line

$$= a \left( \frac{2\theta}{\varphi - 2\theta} \right) \quad \text{cm}$$

which determines the proper location of the photographic plate used for detection. The mass, or more correctly  $m/e$ , is determined by the location of the corresponding ion-beam image on the plate with respect to one or more accurately known masses. The relative abundance of isotopes is determined from the comparative intensities of the isotope images.

This type of mass spectrograph is referred to as velocity focusing since ions with identical  $m/e$  but slightly inhomogeneous energies are focused at a point. Ions entering the electric field at slightly different angles, however, cannot be focused, and the ion-beam currents are therefore exceedingly small as a result of the beam attenuation necessary for a high order of collimation.

**9.2. Dempster Mass Spectrograph.** With the original Dempster instrument [3] the separation of ions with different values of  $m/e$  is accomplished by deflection of the ions through an angle of 180 deg in a uniform magnetic field. The ions formed in an ionization chamber  $I$  (Fig. 61) by electron bombardment are accelerated and focused by voltages applied to the slits  $S_1$ ,  $S_2$ ,  $S_3$ , imparting a final velocity given by

$$v = \left( 2V \frac{e}{m} \right)^{1/2} \quad \text{cm/sec}$$

where  $V$  = voltage difference between slits  $S_1$  and  $S_3$

$e$  = ion charge

$m$  = ion mass

In the uniform magnetic field the ions follow circular trajectories normal to the field and with uniform linear and angular velocity at a radius of curvature determined by

$$R = \frac{c}{H} \left( 2V \frac{m}{e} \right)^{1/2} = \frac{1}{H} \left( 2.07 \frac{mv}{e} \times 10^4 \right)^{1/2} \quad \text{cm}$$

where  $H$  = magnetic field strength, gauss

This form of spectrograph possesses some advantage in utilizing slightly divergent ion beams; hence, somewhat greater beam currents are available than in velocity-focusing instruments. It possesses the disadvantage,

however, that energetically inhomogeneous ion beams cannot be properly focused. The ion source, therefore, must provide ions of as nearly homogeneous energy as possible when the greatest accuracy and resolution is required.

The focus at the 180-deg position where the collecting electrode is placed is not a line image but has a natural width  $\delta$  given by  $\delta = R\alpha^2$ , where  $\alpha$  is one-half the angular beam divergence in radians at the source. This places a limit on the separation of two values of  $m/e$  with nearly equal magnitude, or alternatively, determines the maximum allowable angle of beam divergence for a required resolution, since the separation of the two foci must be greater than the image width, or approximately

$$\alpha^2 < 2$$

Assuming the maximum geometrical resolving power to be defined as the smallest difference in mass  $\Delta m$  for which the images are just separated, it is then given by the expression

$$\frac{m}{\Delta m} = 2\alpha^2$$

This assumes the ion beam to be perfectly homogeneous in velocity; if it is not, a term involving  $\Delta V$  must be added.

**9.3. 60-Degree (Nier-Type) Mass Spectrometer.** A mass spectrometer employing a uniform 60-deg magnetic field has been developed into a highly practical instrument by Nier for routine gas analyses and isotope-abundance measurements [9,23]. Aside from its source and electrical components, the chief advantage of this type of instrument is in the relatively small magnet required to maintain the wedge-shaped field. On the other hand, since an electrostatic analyzing field is not used, this type of geometry does not, strictly speaking, provide either velocity or directional focusing and its resolution therefore depends on the efficiency of the ion source and the degree of collimation of the ion beam.

The geometrical arrangement of the source, magnetic field, and ion collector is shown in Fig. 62. Some adjustment must be provided for moving the magnet along the axis *A-A* since the fringing field of the magnet alters somewhat the exact geometry. In the uniform portion of the field, the ion trajectory radius is given by

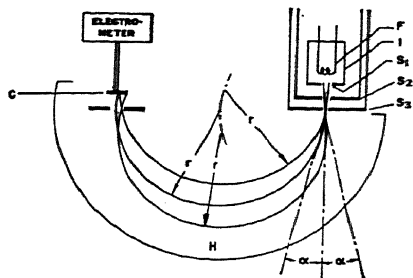


FIG. 61. Dempster mass spectrograph.

$$R^2 = 2.07 \frac{m}{e} \frac{V}{H^2} \times 10^4 \quad \text{cm}^2$$

where  $m$  = ion mass, mass units

$e$  = ion charge, units of electronic charge

$V$  = accelerating potential, volts

$H$  = magnetic field strength, gauss

With a radius of roughly 15 cm and ion source and collecting slit widths of the order of 0.2 mm, resolution of adjacent mass peaks is possible up to mass 400.

**9.4. Mattauch Double-focusing Spectrograph.** A comprehensive study of ion optics by Herzog [4] demonstrated that with appropriate combinations

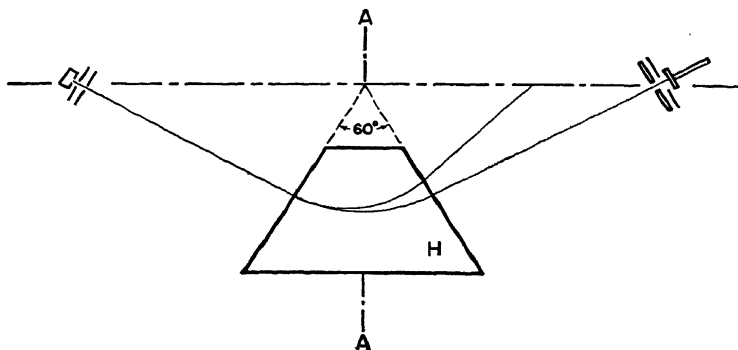


FIG. 62. Mass spectrometer using 60-deg magnetic deflection (Nier type).

of electric and magnetic fields it is possible to achieve both velocity and directional focusing. Where the highest resolution and accuracy are required this is essential since ion sources do not provide ion beams which are strictly homogeneous either in energy or direction. Following these principles Mattauch and Herzog [5,6] constructed a mass spectrograph that provides directional and velocity focusing over the entire mass range. As shown in Fig. 63, partially collimated ions from the source pass through an electric field between condenser plates  $E$  which form an arc of  $31^\circ 51'$ . Again collimated by slit  $S_3$ , the ions enter the magnetic field where they are deflected through an angle of  $90^\circ$ . Directional focusing occurs in the electric field which forms a velocity spectrum at its focal plane, and velocity focusing takes place in the magnetic field.

The voltage applied across the electrodes of the electrostatic analyzer depends upon both the geometry and the voltage through which the ions are initially accelerated at the source. For ions that traverse the central path  $C$  through the analyzer, the required voltage is given by the expression



$$V = 2V_o \log \frac{r_2}{r_1} \quad \text{volts}$$

where  $V_o$  = ion accelerating voltage at source

$r_1$  = radius of inner electrode

$r_2$  = radius of outer electrode

The axial distance  $a$  from edge of the magnetic field where the ions enter to the point of focus (see Fig. 63) is given by

$$a = \frac{c}{H} \sqrt{\frac{2mV_o}{e}} \quad \text{cm}$$

where  $H$  = magnetic field strength

$m$  = mass of particle

$e$  = charge of particle

$c$  = velocity of light

All masses are focused on a plane making a 45-deg angle with the axis at the entrance point of the magnetic field. The distance of the focal point

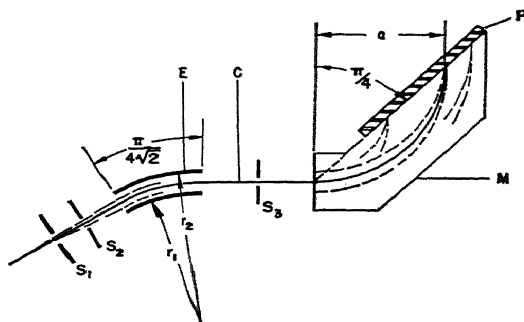


FIG. 63. Mattauch double-focusing mass spectrograph. [*J. Mattauch and R. Herzog, Phys. Rev.*, **50**, 617 (1936).]

along this plane for a particular value of  $m/e$  is then

$$b = a \sqrt{2} \sqrt{\frac{mV_o}{e}} \quad \text{cm}$$

Hence, the mass scale varies as  $\sqrt{m}$  and can be readily established from accurately known masses.

The dispersion, assuming  $e$  to be constant, is given by

$$\frac{db}{dm} = \frac{1}{H} \sqrt{\frac{V_o}{em}}$$

and the resolving power is given by the expression

$$\frac{m}{\Delta m} = \frac{a}{2s_1}$$

where  $s_1$  = source slit width

$\Delta m$  = smallest detectable mass difference at mass  $m$

**9.5. Dempster Double-focusing Spectrograph.** A directional and velocity focusing arrangement of electrostatic and magnetic fields was proposed by Dempster [27] in which the ion beam is deflected through an angle of 90 deg

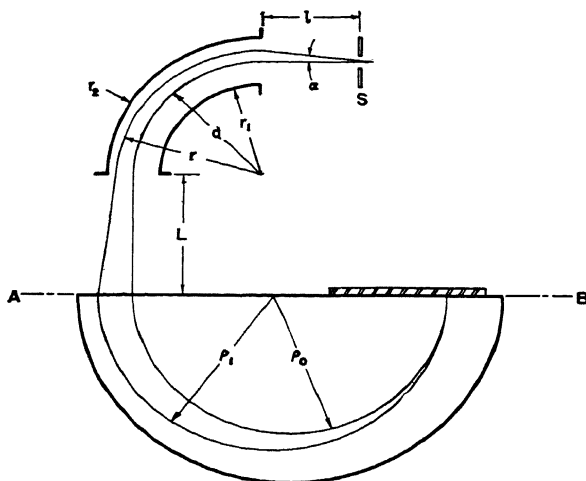


FIG. 64. Dempster double-focusing mass spectrograph.

in a cylindrical electric field and through 180 deg in the uniform magnetic field. The geometrical arrangement of the fields and ion paths is shown in Fig. 64.

Ions of slightly different energies leaving the source  $S$  through the exit slit at a distance  $l$  from the electrostatic analyzer are focused into a velocity spectrum at a distance  $L$  after traversing the analyzer. The potential  $V_e$  across the analyzer plates is related to the energy or accelerating potential  $V_0$  of ions that traverse the central path of radius  $a$  by the expression

$$V_e = 2V_0 \log \frac{r_2}{r_1} \quad \text{volts}$$

Those ions, however, which leave the source at an angle  $\alpha$  and a velocity slightly different from  $v_0$ , i.e.,  $v_1 = v_0(1 + \beta)$ , move on circular trajectories with radii of curvature given by

$$r = a \left( 1 + \frac{\alpha}{2} \sin \frac{\pi}{\sqrt{2}} + \beta + \frac{\alpha l}{a} \cos \frac{\pi}{\sqrt{2}} \right) \quad \text{cm}$$

The relation between the source distance  $l$  and the local plane distance  $L$  is

$$lL - (l + L) \frac{a}{\sqrt{2}} \cos \frac{\pi}{\sqrt{2}} - \frac{a^2}{2} = 0$$

and the velocity dispersion in the focal plane in terms of linear units is

$$d = \beta a \left( 1 - \cos \frac{\pi}{\sqrt{2}} + \frac{L}{a} \sqrt{2} \sin \frac{\pi}{\sqrt{2}} \right) \quad \text{cm-volts}$$

To achieve complete velocity focusing, the velocity dispersion of the magnetic field must compensate exactly for that produced by the electrostatic analyzer. Since the edge of the magnetic field coincides with the focal plane of the analyzer and  $\rho_1 = mv_1/eH$ , this condition is satisfied when  $d = 2\beta\rho_0$ . This condition is exactly met for only one radius of curvature in the magnetic field, and all other radii focus imperfectly at the 180-deg position and hence exhibit progressively wider images with greater distance from the focused value of  $e/m$ . The widths of these foci are given by the expression

$$\delta = \frac{d(\rho_0 - \rho_1)}{\rho_0}$$

It is apparent that when a limiting resolution is required the final focal image width can be made smaller by reducing the dispersion  $d$  with a defining vane placed behind the focal plane of the electrostatic analyzer, *i.e.*, in the plane  $AB$ , Fig. 64.

**9.6. Bainbridge-Jordan Double-focusing Mass Spectrograph.** Directional and velocity focusing is accomplished in a mass spectrograph developed by Bainbridge and Jordan [7], first by deflection of ions in an electrostatic analyzing field through an angle of  $\pi/\sqrt{2}$ , and then in a uniform magnetic field through a mean angle of  $\pi/3$  radians. With the geometrical arrangement used (Fig. 65) the small dispersion in velocity accompanying directional focusing for ions of a particular value of  $m/e$  is canceled by the velocity focusing in the magnetic field. The focal plane over a large mass range is not strictly flat, but over a considerable range about the exactly focused value of  $m/e$  it is sufficiently flat to allow accurate comparison of both mass and abundance. A considerable advantage inherent in this design of spectrograph is derived from the accurately linear mass scale for a broad range about the exactly focused  $m/e$ . In some instances this is highly desirable since it greatly facilitates accurate comparison of masses.

The relation between the voltage applied across the condenser plates of

radius  $\rho$  is given by

$$\rho = \frac{1}{\gamma} \left( v_o^2 + \frac{E^2 c^2}{H^2} - \frac{2Ec}{H} v_o \cos \theta \right)^{1/2}$$

where  $\gamma = \frac{eH}{mc}$

$v_o$  = initial velocity of ion

The resolving power for this type of instrument is given by

$$\frac{m}{\Delta m} = \frac{b}{\Delta b}$$

Ions of different  $m/e$  can be focused on a single collecting electrode by altering the accelerating potential  $V_o$  and the deflecting potential  $V$  ( $= Ed$

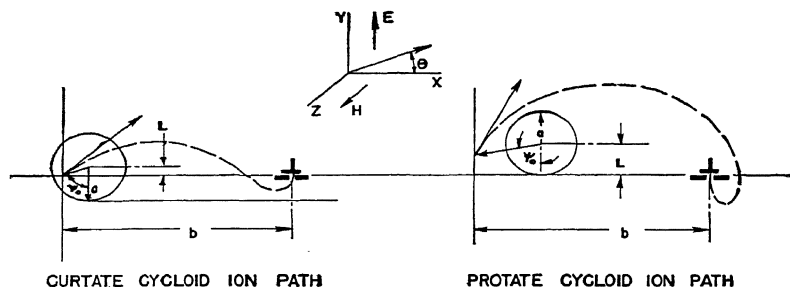


FIG. 66. Trochoidal ion trajectories in crossed electric and magnetic fields. [W. Bleakney and J. A. Hipple, *Phys. Rev.*, **53**, 521 (1938).]

where  $d$  is separation of the condenser plates) in direct proportion, i.e.,  $CV = V_o$  where the constant  $C$  depends upon the design parameters of the instrument. When this condition is fulfilled, ions of any  $m/e$  traverse the same path for the proper values of  $V$  and  $V_o$ .

If the design parameters are chosen to make  $\rho < a$ , the ion trajectory is a curtate cycloid. If  $\rho > a$ , the ion trajectory is that of a prolate cycloid (see Fig. 66).

**9.8. Ion Sources.** The ion sources used in mass spectrographs are either high-voltage discharges or bombardment by low-energy electrons emitted from a hot filament. Both have been widely used, and the choice depends to some extent on the application intended for the mass spectrograph. There is some advantage in using the spark-discharge source for solid materials since the electrodes can be coated with or made of the sample material and little difficulty is encountered in producing ions. Furthermore, the construction and operation of this kind of source is usually simpler than hot-filament sources. On the other hand, filament sources regulated with appropriate circuits give exceedingly stable and reproducible operation over

a long period of time and probably produce ions with smaller spread in energy. Particular advantage is gained for the analysis of gases and vapors with these sources since very small quantities need be used for complete quantitative analyses.

*a. High-voltage Discharge Source.* Ions may be produced in high-voltage sources either by spark discharge in a vacuum chamber or by a steady discharge in an atmosphere of gas at low pressure. The first type of discharge is frequently convenient for the direct ionization of metallic substances. It avoids, among other things, the use of a discharge gas which would produce a spectrum of its own. The sample material can be used as the high-voltage electrode, or when this is impracticable, it can be inserted into a hollow electrode in some usable form, *i.e.*, as a stable salt, an alloy, or a mixture. Suitable high-voltage electrode materials include any of the metals that are stable in a vacuum at elevated temperatures. The material used, however, should not produce ions with values of  $m/e$  near similar values expected from the sample unless they can be used for a comparison spectrum. The discharge takes place in a small gap between the two electrodes which are connected to a tesla coil or some similar high-voltage spark device. Some of the ions that are formed drift to the edge of the discharge in the gap where they then fall through an accelerating potential maintained between the discharge electrodes and a third electrode. This voltage determines the final energy of the ions passing through the analyzing fields.

The second type of high-voltage discharge takes place between a grounded cathode and a high-voltage anode in an atmosphere of gas at low pressure. Satisfactory operation is usually obtained with a potential of approximately 15,000 volts. Unless the gas itself is to be measured, either neon or argon is usually used. Ions formed in the discharge column are accelerated toward the cathode in the electric field of that part of the discharge known as the cathode fall which extends nearly to the anode or to within a distance from it determined by the space-charge sheath thickness. A portion of the ion beam emerges from the discharge tube through a slot in the cathode and is then collimated by an appropriate arrangement of slits, as shown in Fig. 68.

Solid substances that are to be analyzed may be placed on or near the cathode, or in the case of some metallic elements, the cathode can be made wholly of the sample material. Otherwise it is used in the form of a suitable compound, such as a halide salt if it has a vapor pressure greater than  $10^{-3}$  mm Hg at several hundred degrees centigrade. In general, the combined effects of heating and sputtering by positive ions striking the cathode will ensure an ample concentration of sample material in the discharge for the production of ions.

*b. Sources with Electron Emission from a Filament.* This type of source is the most widely used at the present time because of its ease of control,

stability, adaptability to routine analyses of gases and vapors, and the small energy spread of the ions it produces.

The general scheme of this type of source is illustrated by the example shown in Fig. 69. The sample material is admitted to a small closed ioniza-

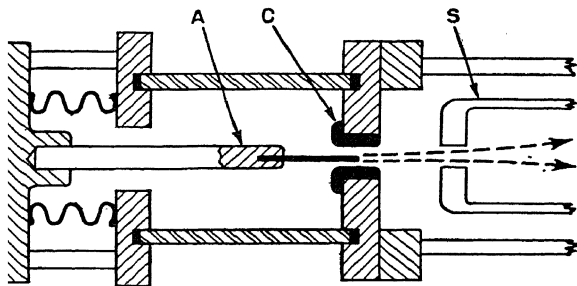


FIG. 67. Spark discharge ion source. Ions formed in the discharge between the end of the anode *A* and the wall of the cathode *C* are accelerated and focused through slit *S*. [*A. E. Shaw and W. Rall, Atomic Energy Commission Report, MDDC-45 (1946).*]

tion chamber whose dimensions are of the order of a centimeter. Ionization is produced by a stream of electrons which enter through a collimating slot at one end and traverse the chamber to be collected at the opposite side by an anode. A fraction of the ions that are produced diffuse out through a slit

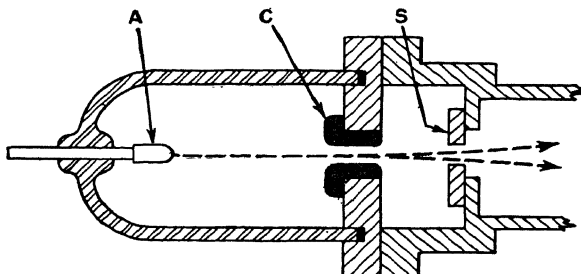


FIG. 68. High-voltage gas-discharge ion source. Ions are formed in the gas discharge maintained between the anode *A* and hollow cathode *C*. Those ions passing through the cathode are then collimated by slit *S* before entering the analyzing fields. Material to be analyzed may be introduced as a gas, or if solid, may be deposited on the cathode. [*K. T. Bainbridge and E. B. Jordan, Phys. Rev., 50, 282 (1936).*]

in one side of the chamber and are then accelerated in an electric field maintained by an appropriate slit or electrode system.

The filament may be made of tungsten or tantalum and used in the form of a helix or a short thin ribbon, usually about 0.001 in. thick. Heating current for filament cross sections commonly used is about 5 amp, alternating

current or direct current. Collimation of the electron stream is obtained with the aid of small electro- or permanent magnets which provide a field of at least several hundred gauss in the region of the ionization chamber. Electrons emitted from the filament move freely along the lines of force but have low mobility in a transverse direction. On the other hand, ions, because of their greater mass, can cross the field with relative ease when leaving the chamber. Electrons, therefore, leave the filament only in the direction of

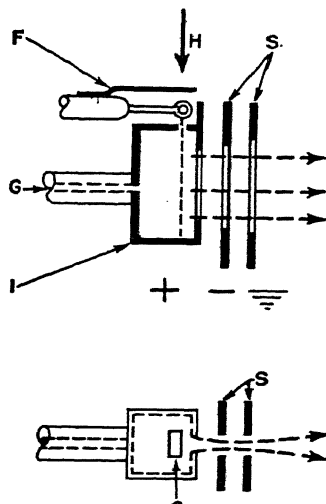


FIG. 69. Simple ion source. Gas introduced through the tube *G* is ionized in the chamber *I* by the stream of electrons (vertical broken line) emitted from the filament *F* maintained at 75 to 150 volts negative with respect to *I*. The electron stream is defined by the magnetic field *H* and collimating slot *C*. Ions that diffuse to the exit slit are accelerated and focused by slits *S*. [*W. Siri, Rev. Sci. Instruments*, **18**, 540 (1947).]

the anode or electron catcher to which they are accelerated by a potential of about 100 volts. In nearly all cases, the total electron emission is less than 1 ma. Of the fraction of electrons that enter the chamber through the collimating slot, a large proportion either is lost to the walls by repeated collisions with gas molecules and ions or is captured by ions. The remaining electrons pass through the chamber to be collected at the anode. Filaments are operated emission limited and require good thermal and emission regulation to ensure stable and consistent operation since the ion-beam current is a rather sensitive function of the emission current.

The slit system through which the ion beam passes serves both to accelerate the ions to their final velocity before entering the analyzing fields and to

focus the beam to the smallest possible angular spread. Numerous combinations of electrodes have been used to achieve these effects, and two examples are shown in Figs. 69 and 70.

Permanent gases and vapors of volatile liquids are introduced directly into the ionization chamber through capillary tubes in which the flow is

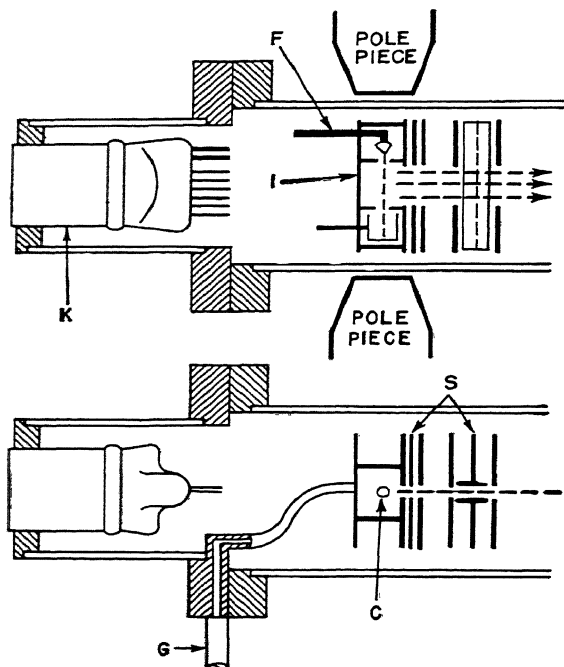


FIG. 70. Ion source developed by Nier. Gas introduced at *G* is ionized in the chamber *I* by stream of electrons emitted from filament *F*. The electron stream is defined by slot *C* and by the magnetic field produced by permanent magnets. Ions are accelerated and focused into a narrow beam by slit system *S*. Electrical leads are brought into the vacuum tube through Kovar seal *K*. [A. O. Nier, *Rev. Sci. Instruments*, **18**, 398 (1947).]

regulated by a valve or throttling device [9–11]. Solid substances can be analyzed usually by one of several methods listed below.

**Sublimation or Distillation:** Some metals, e.g., lithium, calcium, aluminum, and magnesium, and certain compounds of less volatile elements, usually in the form of solids, can be distilled from a hot filament or furnace in or near the ionization chamber.

**Sputtering:** Substances that sputter readily will frequently give a measurable spectrum when a small quantity is inserted in the ionization chamber



near the exit slit. This effect sometimes produces an undesirable background spectrum when light elements are used in the construction of the ionization chamber, *e.g.*, glass, tin, or aluminum.

*Carriers:* Heavy metals sometimes can be transported by a stream of chlorine or fluorine which forms halides of the metals. Most of these halides, however, condense at room temperature, and it is therefore necessary to maintain the sample and gas at an elevated temperature until it enters the ionization chamber.

**9.9. Detector Requirements.** The accuracy of measurements of relative abundance of isotopes or of the constituents of a sample under analysis depends largely upon the sensitivity, linearity, and stability of the device used to measure the focused ion beams of different  $m/e$ . This places severe requirements on the detector since the ion currents may range in magnitude from less than  $10^{-16}$  up to  $10^{-9}$  amp. Hence, in addition to a high order of sensitivity, the response of an accurate mass spectrograph should be reproducible and linear over a range of beam intensity at least of the order of  $10^3$ .

**9.10. Photographic Plates.** The applications that have required the greatest accuracy have been determinations of isotope mass (relative to  $O^{16} = 16.00000$ ). In nearly all such determinations photographic plates have been employed for ion detection. Since they are essentially an integrating device, exceedingly small ion currents can be detected by prolonged exposure, and fluctuations in the current will not affect the accuracy of measurements of relative beam intensities. Both abundance and mass measurements are made from a microphotometric recording of the focal images on the photographic plate. The relative area under the intensity curve of a focal image gives directly the relative abundance, and the distance between maxima gives the relative masses in the appropriate mass scale for the instrument. When a wide range of beam intensities is analyzed, the response of photographic plates cannot be considered linear. In such cases it is nearly always necessary to make several exposures of different values of integrated current on the same plate. For the greatest accuracy, calibration of the emulsion response of the plates should be carried out for a wide range of integrated currents and ion velocities. Since it is known that the emulsion darkening, *i.e.*, number of grains reduced, varies markedly with the ion velocity, this latter effect may be appreciable over the length of the spectrum usually photographed at any one time.

Photographic emulsions should be uniform and show little or no fog in development. Further, it is important in abundance measurements that no solarization occur around the most dense images.

**9.11. Electrical Devices.** The earliest electrical devices for detecting ion beams in mass spectrographs were electroscopes and electrometers. These instruments are current-integrating devices and hence unsuited to rapid

mass analyses. They have, therefore, been wholly replaced by vacuum-tube amplifiers which provide the requisite sensitivity and also permit less sensitive meters and recording instruments to be used. Most frequently, the first stage of an amplifying system is an electrometer-type vacuum tube coupled directly to the ion-collecting electrode of the mass spectrometer. These tubes usually contain four elements and are designed to draw very small grid currents and to operate with plate potentials of only 7 to 20 volts. The input signal is applied to the second grid, or what is normally the screen grid. Several electrometer tubes with very high sensitivities and suitable characteristics have been designed especially for this type of application. The FP-54 [12] is the most sensitive of these tubes and can be used for continuous indication of ion currents as small as  $10^{-15}$  amp. Several other electrometer tubes are also available such as the Vx-41 [13] and the 38, 954, and 959 [19] which, though somewhat less sensitive, are, under some conditions, more stable.

The output from the electrometer tube may be used directly to drive a sensitive galvanometer, or the signal can be further amplified by successive stages of d-c amplification.

The signal voltage driving the grid of the electrometer tube is derived from the voltage  $E$  developed across a grid resistor of the order of  $10^8$  to  $10^{11}$  ohms through which the ion current passes to ground. The maximum sensitivity that can be achieved in circuits of this type is limited by the average voltage  $\bar{e}$  produced in the resistor by thermal agitation. This is given [14-16] approximately by the relation

$$\bar{e}^2 = 4\pi TR\Delta f$$

where  $T$  = absolute temperature

$\Delta f$  = frequency band passed by amplifying system

Assuming that the minimum detectable ion current  $i_0$  is that which produces across the resistance  $R$  a voltage equal to  $e$ , then

$$i_0 = 1.29 \times 10^{-10} \frac{(\Delta f)^{1/2}}{R}$$

Although it is apparent that the signal-to-noise ratio can be improved by using a high input resistance, values of  $R$  greater than  $10^{11}$  ohms are difficult to obtain and still more difficult to maintain at a constant value. Variations in temperature and particularly in surface resistance are sometimes difficult to control and, together with variation in the effective resistance with signal voltage, may lead to drift and transient changes in the sensitivity of the instrument. More important still is the tendency of some resistors to polarize, thus introducing a long signal decay time. The band-pass fre-

quency can be made very small only when the ion current is changed very slowly. If the mass spectrum is scanned as it is in nearly all automatic recording mass spectrometers,  $\Delta f$  must be sufficiently wide to allow the galvanometer or recording device to follow faithfully the rapid increase and decrease in ion current as each mass peak sweeps past the collecting electrode; otherwise resolution is lost because adjacent mass peaks overlap, or alternatively, time and labor are sacrificed in waiting for the signal voltage to decay to the base-line level. For the same reason it is also important to reduce to a minimum the shunting capacitance of the input to the electrometer tube by making the lead from the collecting electrode as short as possible

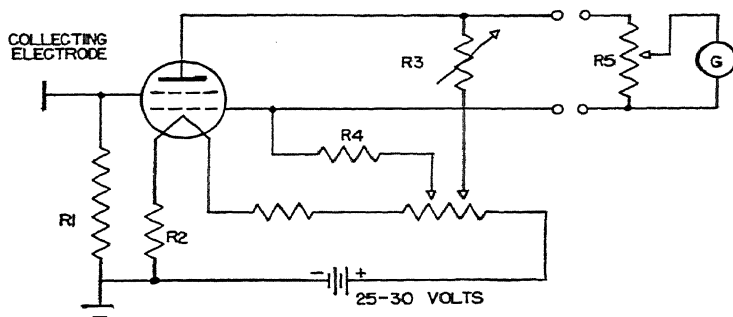


FIG. 71. Balanced-bridge electrometer circuit. Resistance  $R_1$  is usually of the order of  $10^{10}$  ohms. If an FP-54 electrometer tube is used, the filament current is about 90 ma and  $R_2$  is chosen to give a filament bias of about  $-4$  volts. The other circuit constants are chosen to give the required grid and plate voltages and adjusted to give zero galvanometer current for zero input signal. The total resistance  $R_3 + R_4 + R_5$  across the galvanometer should equal the critical damping resistance for the galvanometer. If an inverse feedback d-c amplifier is coupled to the output,  $R_1$  is connected to the feedback return lead instead of to ground. Except for the galvanometer and the divider  $R_5$ , the entire electrometer circuit should be carefully shielded and shock mounted. (See also reference 22.)

and by using shields of large diameter, thus making  $\Delta f$  sufficiently wide to pass the audio-frequency Fourier components of a slow pulse.

Considerable care must be exercised in mounting and shielding electrometer tubes since their high sensitivity also makes them highly microphonic and extremely sensitive to stray electric and magnetic fields. Drift, transients, and microphonics are best avoided by carefully shock mounting the tube in a rigid brass or copper can which is then evacuated to prevent sound transmission and changes in the surface resistance of both the grid resistor and the surface between the tube terminals, mainly by the removal of water vapor.

The conventional electrometer circuit is a balanced-bridge network such as that shown in Fig. 71. For the FP-54 the power supply is 25 to 30 volts,

depending upon the circuit values used, and it may be supplied from batteries or an electronic supply. If the latter, it should be extremely well filtered and very highly regulated in order to avoid drift and short period transients. Extreme care must be taken in the assembly of such circuits to eliminate contact potentials, acid soldering flux, short leakage paths, etc., and all

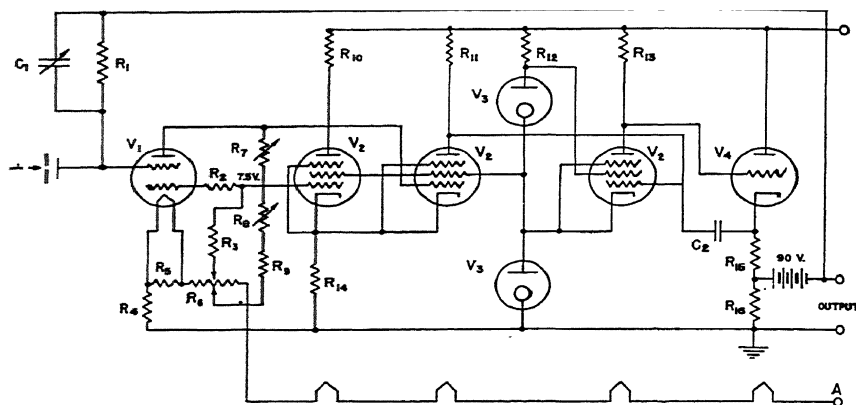


FIG. 72. Typical example of current amplifiers that are now used for measuring steady currents as small as  $10^{-15}$  amp. The negative feedback, d-c amplifier shown here was developed by Nier especially for use with mass spectrometers. The power supply voltage for the plates is 225 volts, and the voltage at *A* is adjusted for a filament current of 150 milliamperes. Precision wire-wound resistors are indicated by PWW. [Redrawn from A. O. Nier, *Rev. Sci. Instruments*, **18**, 398 (1947).]

$R_1 = 4 \times 10^{10}$  ohms, IRC type MG-6  
 $R_2 = 5,000$  ohms, PWW  
 $R_3 = 20,000$  ohms, PWW  
 $R_4 = 40$  ohms, PWW  
 $R_5 =$  shunt to limit filament current in  $V_1$  to 20 ma  
 $R_6 = 12$  steps of 13 ohms each, PWW  
 $R_7 = 200,000$  ohm potentiometer  
 $R_8 = 20,000$  ohm potentiometer  
 $R_9 = 200,000$  ohms, PWW

$R_{10} \ R_{11}, R_{13} = 1$  megohm, 1 watt  
 $R_{12} = 7,500$  ohms, 10 watts  
 $R_{14} = 35,000$  ohms, PWW  
 $R_{15} = 10,000$  ohms, PWW  
 $R_{16} = 15,000$  ohms, PWW  
 $C_1 = 100 \mu\text{fd}$ , variable condenser  
 $C_2 = 0.02 \mu\text{fd}$ , 400 volt condenser  
 $V_1 = V\text{X-41}$  (Victoreen Instrument Co.)  
 $V_2 = 12\text{SJ7}$   
 $V_3 = \text{VR75}$   
 $V_4 = 12\text{J5}$

resistive elements should be wound from zero temperature-coefficient wire. The use of long leads from the output to a galvanometer or recording device is permissible.

The nonlinear characteristics of the electrometer tube over the range of currents ( $10^{-14}$  to  $10^{-10}$  amp) which is ordinarily measured must be accurately compensated. In manual or static measurements the electrometer-gal-

vanometer circuit is used as a null indicator. When a mass peak has been maximized on the galvanometer, an accurately linear voltage from a potentiometer or decade-battery circuit is applied to the bottom of the input grid resistor, deflecting the galvanometer back to the base line. The peaks are then found in terms of millivolts rather than centimeters of deflection and are as accurately measured as the applied back voltage is linear. A more satisfactory method that is rapidly replacing the simple electrometer-galvanometer circuit is found in the use of a high gain inverse-feedback amplifier coupled directly to the output of the electrometer stage. With a d-c amplifier gain of 5,000 to 10,000 and 100 per cent negative feedback to the input resistor of the electrometer circuit, the over-all voltage gain of the system is very nearly 1, but the output voltage is accurately proportional to the input. In addition to its linearity, this type of amplifying system has high inherent stability, usually not found in the simple balanced bridge electrometer circuit, and a response time considerably shorter than most recording mechanisms. An example of this type of circuit, developed by Nier [13], is shown in Fig. 72.

Other detector circuits have been proposed which utilize a-c amplifiers and a pulsed ion beam to provide the necessary audio-frequency signal. In one system this is done by oscillating the ion-beam radius in the magnetic field at a frequency of approximately 200 cps by oscillating the accelerating voltage [10]. The pulse received by the collecting electrode as the beam sweeps past is amplified and observed on an oscilloscope in which the linear sweep is synchronized with the beam-sweep frequency. Alternatively, after amplification the pulse may be rectified to operate a d-c recording meter. A second scheme accomplishes essentially the same result by pulsing the ion source [18].

**9.12. Mass-spectrometer Recording Systems.** Routine mass spectrometer analyses are now usually carried out by automatically scanning and recording the entire mass range or those portions of the spectrum which may be of interest. The scanning may be done by slowly varying either the electric ion accelerating field or the magnetic field. Some advantage is to be found in the latter method in that the magnetic field varies only as  $m^{1/2}$ , whereas the electric accelerating field varies directly with  $m^{-1}$  and, therefore, often involves an inconveniently large range in voltage. A serious disadvantage of voltage scanning sometimes arises from voltage discrimination, an effect that is important primarily in isotope analyses where the increased accelerating voltage for smaller mass favors the observed beam currents of the lighter components of the isotopes. Whether or not this effect is present in all types of ion sources is not yet certain, but when it is present it appears to be an involved function of the source geometry, the electric field strength, and the accelerating voltage difference between any two masses.

Among the various recording mechanisms that have been developed, the most straightforward is direct recording of the galvanometer light beam on a moving strip of photosensitive paper. With several identical galvanometers connected in series and each shunted for a different sensitivity, a wide range of ion currents can be recorded on a narrow paper strip since the shunts or sensitivities can be chosen so that at least one galvanometer trace remains on the paper for the largest expected beam currents. More complex recording systems now in use employ standard recording instruments, usually recording potentiometers such as the Speedomax.\* Scale contraction in these instruments is accomplished by automatic shunt selection which provides a stepwise sensitivity approximating a logarithmic scale over the width of the recording paper [20]. A further modification of the automatic shunt-selection system enables the top of a peak to be recorded on a linear scale while a large portion of the center of the peak is contracted by a non-linear scaling factor which depends upon the amplitude of the peak [21].

**9.13. Mass-spectrometer Errors.** Relative mass-abundance measurements made with a mass spectrometer are subject to many possible errors, some of which are inherent in the instrument and others in the isotopic species themselves. The observed ion currents of several isotopes do not always represent exactly the mass abundance but must be corrected for one or more of the effects that lead to errors. The more important sources of errors are described below.

*a. Inlet-sample Flow Rate.* The lighter of two molecules flows through the leak or capillary at a higher rate than the heavier component in accordance with the laws of diffusion. If the system allows free molecular flow, *i.e.*, if the mean free path is very much greater than the capillary radius and the pressure gradient through the leak is small, then the flow rate is inversely proportional to the square root of the molecular weight. This effect is particularly important in isotopes of very light mass, *e.g.*,  $H_2$  and  $HD$ .

*b. Pump-out Rates.* As in *a*, the rates at which substances are pumped out of the spectrometer tube favor the lighter masses and, with a liquid nitrogen or oxygen trap, also the condensible vapors. To some extent this may tend to compensate *a*.

*c. Voltage Discrimination.* The ion current varies somewhat with accelerating voltage, *i.e.*, electric field strength at the source. If several mass peaks are measured by voltage scanning, the lighter masses are enhanced progressively by the higher accelerating voltage required to focus them on the collecting electrode. Among the isotopes of light elements the effect can be quite large. It can be avoided, however, by magnetic field scanning or by using several collecting electrodes properly located to receive the ion beams of the different isotopes simultaneously.

\* Leeds and Northrup Company.

*d. Background.* The presence of a background of peaks due to residual gas can introduce serious errors when they appear at mass positions of the less abundant sample components. The only reliable correction is elimination of the background in the region of the spectrum in which mass peaks of the sample components will appear. Except for certain organic compounds, water vapor is the most difficult of the common substances to outgas. Once the system has been exposed to water vapor, a prolonged bake-out with the entire spectrometer tube maintained at an elevated temperature is necessary to remove the water vapor peaks at masses 16, 17, and 18. Also frequently observed are mercury or hydrocarbon peaks due to vapors from the diffusion pump.

*e. Incomplete Resolution.* Peaks of large amplitude may be sufficiently wide at the bottom to overlap the adjacent mass position, thus making a correction necessary for the amount of overlap. This effect arises from poor slit- and magnetic-field adjustment, inadequate collimation, metastable ions [28], or a long time constant in some part of the detecting circuits.

*f. Nonlinearity of Detecting Circuits.* This can be avoided by the use of 100 per cent feedback in the electrometer-amplifier system or by use of a null method.

*g. Instability of Electrical Circuits.* This includes primarily the emission current and voltage and the detecting circuits. If drift and short period fluctuations cannot be avoided, it is necessary to take a sufficient number of readings to give a statistically reliable average.

*h. Overlapping Spectra.* Molecules containing more than one atomic species, each with several isotopes, frequently form combinations of different isotopes but with the same total mass, *e.g.*, carbon dioxide. Determination of the abundance of the separate isotopes or of the molecule is sometimes possible only by analysis of its entire spectrum.

**9.14. Hydrogen.** The hydrogen isotopes are measured in the form of hydrogen gas. Water vapor is impracticable because of the great difficulty in ridding the system of it, the inconsistency of results, and the needless complication by the oxygen isotopes. Attempts to use hydriodic acid have met with consistent failure.

The hydrogen gas sample is usually obtained by reduction of water derived from the original labeled substance. This is most conveniently done by passing the water vapor through a quartz tube containing zinc grains maintained at a temperature of 350°C. The collected hydrogen is then tolepered into a small glass sample bottle for transfer to the gas sampling-system of the spectrometer.

Hydrogen-isotope analysis is somewhat complicated by the formation of  $H_2$  in the ionization chamber under conditions that depend upon the pressure, the source geometry, and the accelerating field strength [24]. The formation of

$H_2$  and other combinations of hydrogen and deuterium depend primarily on the pressure, and under the most general conditions the molecules and masses shown in Table 32 may be observed [24].

TABLE 32. HYDROGEN MOLECULE MASSES

Mass	Molecule	Variation with $P$
1	H	$P, P^2$
2	$H_2$	$P$
	D	$P, P^2$
3	HD	$P$
	$H_3$	$P^2$
4	$D_2$	$P$
	$H_2D$	$P^2$
5	$HD_2$	$P^2$
6	$D_3$	$P^2$

When the concentration of deuterium is small, of the order of 1 per cent or less, it can be assumed that all the deuterium is bound as HD and measurements of only masses 2 and 3 are necessary. Mass 3 is the sum of HD and  $H_3$ , but since  $HD \sim P$  and  $H_3 \sim P^2$ , by plotting the observed values of the ratio

$$\frac{(HD + H_3)}{H_2} = \frac{\alpha P + \beta P^2}{\gamma P}$$

where  $\alpha, \beta, \gamma$  = constants of proportionality against the pressure, or against  $H_2$  to which the pressure is proportional, a straight line is obtained. By extrapolating the line to zero pressure ( $H_2 = 0$ ) the intercept on the ordinate gives directly the ratio  $\alpha/\gamma = HD/H_2$ . When the concentrations of hydrogen and deuterium are comparable, a more complete analysis is indicated.

**9.15. Carbon.** Carbon is usually introduced into the mass spectrometer as carbon dioxide. Its preparation from the sample substance is straightforward. Carbon dioxide driven off by combustion of the substance is absorbed in sodium hydroxide and the solution neutralized by addition of ammonium nitrate or ammonium chloride in excess. Barium chloride is then added in excess to precipitate the carbonate which is filtered or centrifuged, washed with water, and dried in an oven at 110 to 135°C. If the initial alkali solution is weak, *i.e.*,  $\leq 1N$ , barium chloride can be added directly. A few milligrams of dried barium carbonate are transferred to a single-ended quartz microcombustion tube which is connected to the sampling system. After preliminary heating under vacuum to drive off



occluded gases, the quartz tube is heated to 1100°C in a gas-oxygen flame until the barium carbonate is completely dissociated. A detailed discussion of carbon-isotope chemistry will be found in reference 25.

Determination of the ratio  $C^{13}/C^{14}$  is obtained directly from the mass peaks 44 and 45 corresponding to  $C^{12}O^{16}O^{16}$  and  $C^{13}O^{16}O^{16} + C^{12}O^{16}O^{17}$ , respectively. The contribution of  $C^{12}O^{16}O^{17}$  to mass 45 is usually negligible. Masses 12 and 13 can also be used, but the beam intensities are considerably smaller than for the molecular ions. They are not, however, difficult to measure, and more important, they are not affected by the oxygen isotopes. In terms of the ratio  $R$  of the intensity of  $C^{12}$  (mass 12 or 44) to  $C^{13}$  (mass 13 or 45) the atom per cent concentration of  $C^{13}$  is

$$\% C^{13} = \frac{100}{R + 1}$$

where  $R = C^{12}$  reading/ $C^{13}$  reading

**9.16. Nitrogen.** The relative abundance of the nitrogen isotopes 14 and 15 in a sample substance is determined from measurements of the nitrogen molecule masses of 28, 29, and 30. The preparation of the nitrogen from the organic sample material requires first its reduction to ammonia and then oxidation of the ammonia to free  $N_2$  by hypobromite [26]. The ammonia obtained from the sample substance is contained in 0.05*N* hydrochloric acid which is boiled down to drive off dissolved gases and then transferred to a nitrogen generating system. With the system evacuated, hypobromite is added to oxidize the ammonia according to the reaction



The nitrogen is toeplered into a small sample bottle equipped with a stopcock and transferred to the spectrometer sampling system.

For  $N^{15}$  concentrations of the order of 1 per cent and less, only masses 28 ( $N^{14}N^{14}$ ) and 29 ( $N^{14}N^{15}$ ) need be measured. The probability for formation of  $N^{15}N^{15}$  is negligibly small. The atom per cent of  $N^{15}$  in the sample is

$$\% N^{15} = \frac{100I^{29}}{2I^{28} + I^{29}} = \frac{100}{2R + 1}$$

where  $I^{28}$  = ion current of mass 28 ( $N^{14}N^{14}$ );

$I^{29}$  = ion current of mass 29 ( $N^{14}N^{15}$ )

$$R = I^{28}/I^{29}$$

When the concentration of  $N^{15}$  is greater than several per cent, it is necessary to measure masses 28, 29, and 30 since the contribution of  $N^{15}N^{15}$  becomes appreciable. The atom per cent of  $N^{15}$  is then

$$\% N^{15} = \frac{100(I^{29} + 2I^{30})}{2(I^{28} + I^{29} + I^{30})}$$

It is obviously important in measuring nitrogen to avoid air leaks, dissolved or adsorbed air in the solutions, in the sampling system, or in the spectrometer. The presence of air contamination is made evident by the oxygen peak at mass 32. If the ratio of the nitrogen 28 peak to the oxygen 32 peak for normal air has been determined previously for the spectrometer, a correction for a small per cent of air contamination can be made readily from the measured intensity of the  $O_2$  peak which, when multiplied by the N O ratio, gives the required mass 28 correction.

**9.17. Oxygen.** The abundance of the oxygen isotopes in  $O^{18}$ -enhanced samples may be determined from measurements of the oxygen molecule masses 32, 33, 34, etc., or by introducing the oxygen as carbon dioxide and measuring masses 44, 45, and 46. If the concentration of  $O^{18}$  is a few per cent or less, contributions by molecules of the form  $O^{17}O^{17}$ ,  $O^{17}O^{18}$ , and  $O^{18}O^{18}$  are entirely negligible and only masses 32, 33, and 34 are measured. The atom per cent of  $O^{18}$  is then

$$\% O^{18} = \frac{100I^{34}}{2(I^{32} + I^{33} + I^{34})}$$

where  $I^{32}$  = intensity of mass 32

$I^{33}$  = intensity of mass 33

$I^{34}$  = intensity of mass 34

A similar analysis is applied for measurements of oxygen in the form of carbon dioxide. Neglecting the contributions by  $C^{13}O^{16}O^{17}$  and  $C^{13}O^{16}O^{18}$ , masses 44, 45, and 46 give the atom per cent of  $O^{18}$  by the formula above. For high concentrations of  $O^{18}$  it may also be necessary to measure mass 47.

Air contamination is important, but if its percentage is small its contribution to the measured peaks can be corrected. This may be done by measuring the nitrogen 28 peak. If the ratio of the oxygen 32 mass to nitrogen 28 mass for normal air is known for the instrument, the appropriate corrections are made as indicated in the last section for nitrogen.

The oxygen atomic masses 16, 17, and 18 usually cannot be measured accurately because of the background of water-vapor peaks.

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## CHAPTER 10

### GEIGER-MÜLLER COUNTERS

**10.1. General Properties.** Geiger-Müller tubes are a form of high-gain, gas-filled, amplifying diode operated at potentials below the continuous discharge voltage. Their essential function is that of a triggering device in which a voltage pulse is produced by a discharge initiated by an ionizing particle. The accumulation of negative charge, reaching a magnitude of the order of  $10^{-8}$  coulomb, is collected at an anode where it can be detected by appropriate instruments. The usual form of Geiger-Müller tube consists of a cylindrical cathode and a coaxially mounted wire anode sealed in a tube containing one of various possible gas mixtures, usually at reduced pressure. Designs, materials, and gas mixtures of many varieties are used, the choice depending largely on the kind of radiation to be detected and to some extent on special purposes. The tubes commonly used vary in size from 0.3 to 10 cm in diameter and from 2 to 50 cm in length, and for most tubes the anode wire, usually tungsten, is about 0.02 to 0.1 mm in diameter.

The size, duration, and general character of the discharge in a counter tube is independent of the specific ionizing power of the initial particle. Thus an electron and an alpha particle produce the same pulse as observed on an oscilloscope screen although the latter particle produces  $10^3$  to  $10^5$  times as many ion pairs per centimeter of path. One ion pair, if formed in the sensitive region of the tube, is sufficient to trigger the discharge which subsequently involves roughly  $10^{10}$  ion pairs.

The first part of the discharge occurs rapidly. Electrons released by the initial ionizing particle drift rapidly to the central anode wire. In the high electric field close to the wire the electrons acquire sufficient energy between collisions to ionize the neutral gas molecules releasing additional electrons which further contribute to a Townsend avalanche. Thus the initial number of free electrons is increased by a factor of  $10^3$  to  $10^{10}$ . Because of the high mobility of electrons, this part of the process is completed in a microsecond or less. The less mobile positive ions, however, remain as a positive space charge surrounding the anode along its entire length [1,2], and the subsequent behavior of the discharge depends upon the composition of the gas used to fill the counter, more generally, depending upon whether it is a quenching or nonquenching gas.

**10.2. Non-self-quenching Counters.** When a counter tube contains monatomic, diatomic, or certain triatomic gases, the discharge tends to

maintain itself and the counter tube is said to be non-self-quenching or "slow." Following the generally accepted qualitative description of the processes given by Montgomery and Montgomery [3] and other writers [4], the presence of the residual slow-moving positive ion sheath greatly modifies

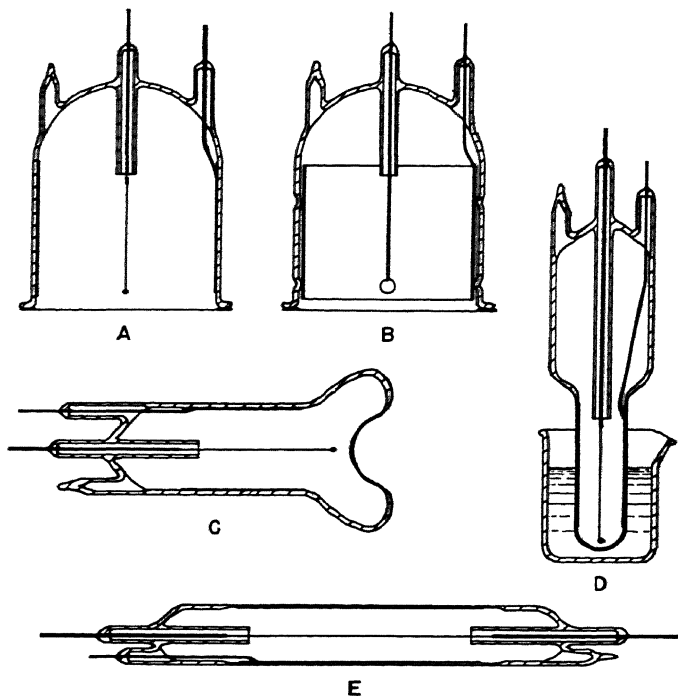


FIG. 73. Typical counter-tube designs.

- A. Bell counter. Wire anode, thin mica window, and chemically deposited silver cathode (beta particles).
- B. Point counter. Ball anode, thin mica window, and alternative inserted copper cylinder cathode (beta particles).
- C. Thin glass window counter (beta particles).
- D. Immersion counter. Thin glass wall, deposited silver cathode. Shown immersed in sample liquid (beta particles).
- E. Cylindrical counter. Thin glass wall and deposited silver cathode for beta particles. Heavy wall and copper mesh cathode for gamma rays.

the electrostatic field gradient surrounding the anode, thus quenching the avalanche by preventing the remaining free electrons from acquiring sufficient energy between collisions to cause further ionization. The positive ions then drift to the cathode in a time of the order of  $10^{-4}$  sec. At the

cathode surface they are neutralized by ejecting an electron from the wall. This is immediately followed by the emission of one or more photons from each atom as it returns to the ground state. For some gases such as argon, helium, and hydrogen, the emitted radiation lies in the ultraviolet and

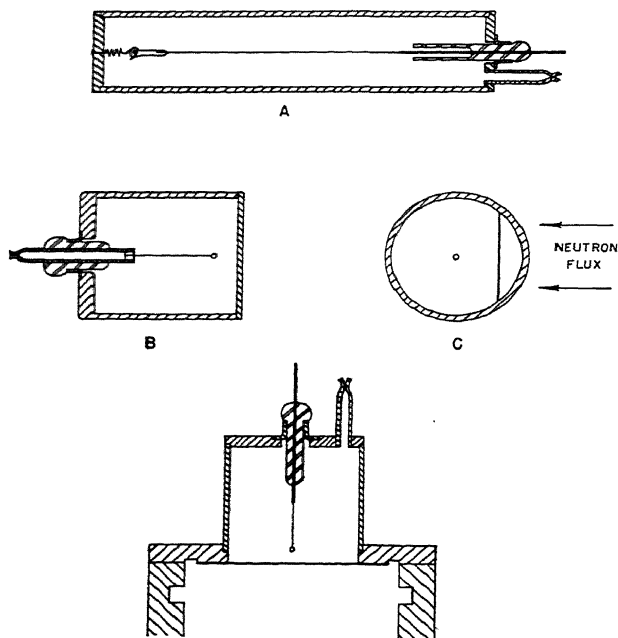


FIG. 74. Easily constructed metal counting-tube designs.

- A. Cylindrical brass or copper tube counter using Kovar seals (cosmic rays, gamma rays, and neutrons).
- B. Copper "bell" counter with Kovar seal pump out and anode lead (gamma rays and neutrons).
- C. Arrangement of radiator in fast neutron counters showing hydrogenous layer coated on a thin platinum plate.
- D. Bell counter with mica window and with mounting for standard sample position (beta particles).

possesses sufficient energy to eject photoelectrons from the counter walls where it is absorbed. Although the photoelectric efficiency is of the order of one ejected electron per  $10^4$  photons, if  $10^{10}$  ions are formed in the discharge process, the probability of electron ejection is high and consequently the discharge is continued. In addition, positive ion bombardment of the cathode at potentials normally used in operating counter tubes frequently

results in the direct ejection of free electrons which may also continue the discharge. A third mechanism observed by Ramsey [5] and others occurs when the initial avalanche is small. Photons emitted by those ions in the avalanche which capture an electron before reaching the cathode can eject photoelectrons which then maintain the discharge in what is observed to be a series of diminishing pulses.

Discharges in tubes containing only mono- or diatomic gases, consequently, will, with certain exceptions, continue so long as the anode potential is maintained. Quenching is accomplished in such tubes only by some external device such as a high resistance, usually of the order of magnitude of  $10^9$  ohms, or by an electronic circuit which reduces the anode potential below the threshold voltage after the initial part of the pulse and until the positive ions are collected. These counters have revolving times of several times  $10^{-4}$  sec when appropriate external electronic quenching circuits are used, but they may be as long as  $10^{-2}$  sec with simple resistance quenching. The minimum resolving time is limited by the positive ion transit time.

**10.3. Self-quenching Counters.** The addition of polyatomic gases such as methane, alcohol, and amyl acetate to counter tubes alters the process in a way to quench the discharge following the Townsend avalanche without the use of external quenching circuits, and such counter tubes are called self-quenching or "fast" counters. The resolving times of such counters is usually of the order of  $10^{-4}$  sec.

The principal function of the polyatomic quenching gas is to prevent further production of electrons following the completion of the initial Townsend avalanche. This is accomplished mainly by reducing photoemission and secondary electron emission from the cathode walls [4].

The effectiveness of polyatomic gases in absorbing photons is due to the diffuse vibration-rotation interaction absorption bands present in the ultra-violet region of their spectrum. Radiation emitted by excited argon, helium, and most other permanent gases lies between 1,020 and 790 angstroms and can, therefore, be absorbed by such molecules as methane and alcohol, which have continuous or band absorption spectra blanketing this region. In addition to absorbing radiation, it is equally important that the quenching-gas molecules do not reemit the absorbed photons but have, instead, a strong tendency toward releasing the excitation energy by photodecomposition. This appears to be valid for all polyatomic gases exhibiting quenching properties.

Secondary electron emission from the cathode is effectively reduced by two mechanisms. (1) The monatomic and diatomic ions formed in the initial ionization process are prevented from reaching the wall by the process of electron exchange with neutral polyatomic molecules with which they collide with a frequency of roughly  $10^2$  to  $10^3$  times per centimeter of path.

This process is possible only when the ionization potential of the quenching gas is less than that of the nonquenching gas [4]. The photons emitted in the process are absorbed in turn by other polyatomic molecules. (2) The quenching-gas ions, formed either by the initial ionizing event or by electron exchange, are neutralized by electron ejection from the wall, but they exhibit a greater probability for decomposition than for ejection of a free electron or for radiative capture. Altogether, the probability of secondary electron production appears to be 1 electron per  $10^{10}$  ions [6,7] and these can be absorbed by the slow-moving residual quenching-gas ions.

The presence in a counter tube of appreciable quantities of negative ion-forming gases such as  $O_2$ ,  $H_2O$ , and the halogens considerably alters the mechanism of discharge and with it the desirable operating characteristics.

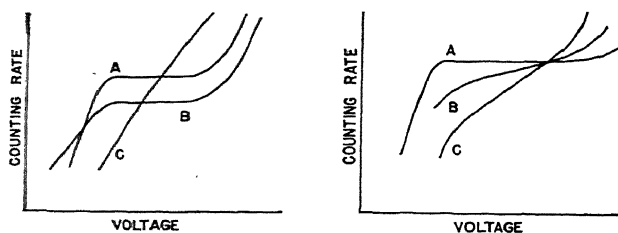


FIG. 75. Effects of gases on G-M plateau. [S. A. Korff and R. D. Present, *Phys. Rev.*, **65**, 274 (1944).]

- A. Pure methane
- B. 1.5 cm argon added
- C. 1.5 cm water added

- A. Pure methane
- B. 1.5 mm air added
- C. 6.0 mm air added

The slow collection time of negative ions compared with electrons tends to prolong the discharge time and frequently leads to the complete disappearance of the Geiger-Müller plateau [4], as seen in Fig. 75.

**10.4. Pulse and Voltage Characteristics.** Voltage pulses as observed with an oscilloscope increase rapidly to a maximum value in a time of the order of 1 microsecond, corresponding to the collection time of the electrons. Following the pulse maximum or pulse rise time, the anode voltage, which has actually been depressed, slowly recovers to normal potential as the residual positive ions are swept out. During the first part of the pulse, referred to as the dead time, the counter is wholly insensitive to a second ionizing event. As the voltage recovers beyond a critical value, a second ionizing event may produce at successively later intervals a larger pulse until, at the termination of the recovery time, a normal pulse voltage is again produced, as shown in Fig. 76. Both the dead time and recovery time appear to have durations of about  $10^{-4}$  sec or less for self-quenching counters.

Voltage characteristics of counters are determined by observing the



counting rate for increasing anode voltage. The important characteristic of a Geiger-Müller, or discharge, counter is the existence of a well-defined plateau voltage region over which the counting rate does not increase appreciably. Normally this region of the curve should be flat and the counting rate should not increase more than 2 to 5 per cent per 100 volts over an

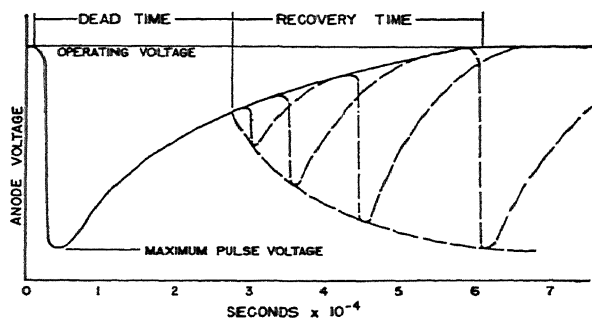


FIG. 76. Counter pulse characteristics. [H. G. Stier, *Phys. Rev.*, 61, 38 (1942).]

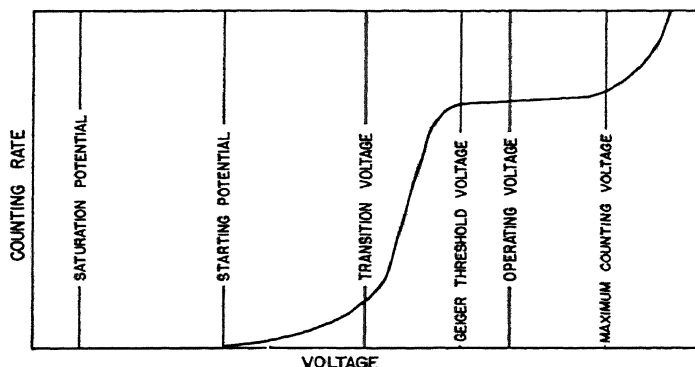


FIG. 77. Counting-rate characteristics of counters as a function of anode voltage.

interval of 100 to 300 volts, although shorter plateaus are satisfactory with well-regulated voltage supplies.

The following voltages are frequently referred to because they are associated with characteristic properties of counter tubes at various potentials (see Fig. 77).

1. Starting voltage. This is the lowest voltage at which counts are observed. It depends largely on the sensitivity of the detecting circuits used.

2. Proportional threshold voltage. This is the lowest voltage at which pro-

portional pulses are observed. It is often identical with the starting voltage, depending upon the sensitivity of the detecting circuits.

3. Geiger-Müller threshold voltage. This is the lowest voltage at which all pulses have the same height.

4. Operating voltage. This is the normal voltage at which a counter is operated. Usually it is in the lower half of the plateau region.

5. Overvoltage. This is the difference between the operating voltage and Geiger-Müller threshold voltage.

In a similar way, certain voltage ranges are distinguished by characteristic behavior of the counter tube. The voltage ranges usually referred to are given below (see Fig. 78).

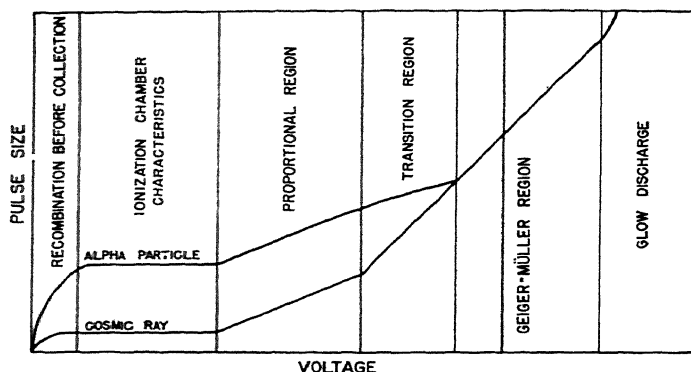


FIG. 78. Ionization produced in counters as a function of anode voltage. [From C. G. Montgomery and D. D. Montgomery, *J. Franklin Inst.*, **231**, 447 (1941).]

1. Region of a few volts above ground potential within which some of the positive and negative ions formed by an ionizing particle recombine before reaching the anode or cathode.

2. Ionization-chamber region, usually in the order of tens of volts. In this region the initial ions formed by the primary particle are collected but do not multiply appreciably. Recombination is negligible.

3. Proportional region. The pulse produced by an ionizing event is proportional to the initial number of ions formed for any fixed operating voltage.

4. Geiger-Müller transition or limited proportionality region. Pulses are no longer strictly proportional, and large pulses may exhibit some of the characteristics of the Geiger-Müller region.

5. Geiger-Müller region. Each event produces a discharge in which pulse size is independent of initial intensity of ionization and the counting rate remains essentially independent of voltage.

6. Continuous-discharge region. This is characterized by corona, glow, and finally, arc discharge.

**10.5. Filling Gases.** The most frequently used filling gas is a mixture of argon (80 to 95 per cent) and alcohol (5 to 20 per cent) at a total pressure of 5 to 40 cm Hg. Although nearly all permanent gases have been used, either alone or in various mixtures, argon is probably the most useful since it has a large cross section for ionization, a sufficiently high ionization potential to allow electron transfer with most polyatomic molecules, does not form negative ions, and is readily available. Many quenching gases other than ethyl alcohol can be used such as ethane, amyl acetate, ethyl ether, and tetraethyl lead. Heavy polyatomic molecules may also be used, and some increase in useful counter life will usually be found because of the greater number of decompositions required to reduce it to a nonquenching gas. However, their use in most counters is undesirable since they lead to a longer resolving time due to the low mobility of heavy ions. Gases that have strong tendencies to form negative ions should normally be avoided [4,9]. These include primarily oxygen, carbon dioxide, water vapor, and the halogens. However, under special conditions they can be used successfully in small amounts [11,14]. It has also been pointed out by Present [36] that chlorine and bromine can under certain conditions be used as quenching gases.

**10.6. Counter-tube Life.** The useful life of self-quenching counters appears to be dependent to some extent upon the volume of gas in the counter [8]. With each discharge approximately  $10^{10}$  ion pairs are formed, and in the process a small fraction of the polyatomic molecules are decomposed into smaller fragments, some of which still retain quenching properties. Ultimately, however, an appreciable fraction of the complex molecules is reduced to free oxygen, hydrogen, carbon, and simple molecules which remain partly as nonquenching gases and partly as an accumulation of crud on the cathode. The counter performance tends to become erratic and finally no longer self-quenching and must be refilled. Assuming that  $10^8$  to  $10^{10}$  quenching gas molecules are decomposed per discharge and an initial number of molecules of approximately  $10^{20}$ , the approximate life of a counter is then of the order of  $10^{10}$  counts. In practice it is found that the smallest counters retain their quenching property for only a few months at best when in constant use, whereas larger counters, with volumes of the order of 75 cc, have useful lives of a year or more under similar conditions. Counters containing only permanent gases should exhibit no change with time, barring leaks or deterioration of the mechanical and electrical properties of the tube itself.

The effective life of self-quenching counters can be prolonged somewhat by operating the counter at a voltage in the lower end of the plateau and by increasing the total gas volume without, at the same time, increasing the sensitive volume in which the discharge takes place. The latter method has

TABLE 33. FILLING GASES USED IN COUNTERS  
 From S. C. Brown, *Nucleonics*, 2, No. 6. 10 (1948).

Gas	Formula	1st ex- citation potential, ev	Ioniza- tion potential, ev	Meta- stable level, ev	Emission or absorption spectrum, angstrom units
Acetone.....	CH <sub>3</sub> COOCH <sub>3</sub>	....	10.1	....	3,300-2,940 abs. bands
Acetylene.....	C <sub>2</sub> H <sub>2</sub>	....	11.6	....	2,400-2,090 abs. bands
Ammonia.....	NH <sub>3</sub>	....	10.5	....	1,620-1,450 abs. bands
Argon.....	A	11.6	15.7	11.5	1,048 emission
Boron trifluoride..	BF <sub>3</sub>	....	....	....	....
Bromine.....	Br <sub>2</sub>	....	12.8	....	1,500 abs. continuous
Carbon dioxide....	CO <sub>2</sub>	....	14.4	....	1,360-600 abs. bands
Carbon disulfide...	CS <sub>2</sub>	....	10.4	....	3,800-1,200 abs. bands
Carbon monoxide...	CO	....	14.1	....	....
Carbon tetra- chloride.....	CCl <sub>4</sub>	....	....	....	4,600-2,300 abs. bands
Chlorine.....	Cl <sub>2</sub>	....	12.8	....	1,500 abs. continuous
Chloroform.....	CHCl <sub>3</sub>	....	....	....	2,200 abs. continuous
Ethyl alcohol.....	C <sub>2</sub> H <sub>5</sub> OH	....	11.3	....	1,633-1,602 abs. bands 1,518 abs. diffuse < 700 abs. continuous
Ethyl bromide.....	C <sub>2</sub> H <sub>5</sub> Br	....	10.24	....	2,850-1,900 abs. continuous < 1,700 abs. diffuse
Ethyl chloride.....	C <sub>2</sub> H <sub>5</sub> Cl	....	....	....	< 1,700 abs. continuous
Helium.....	He	20.5	24.5	19.7	854 emission
Hydrogen.....	H <sub>2</sub>	11.5	15.4	None	1,215 emission
Hydrogen sulfide...	H <sub>2</sub> S	....	10.4	....	1,600-1,190 abs. bands
Krypton.....	Kr	9.9	13.9	9.87	1,236 emission
Mercury.....	Hg	....	10.4	5.43	1,850 emission
Methane.....	CH <sub>4</sub>	....	14.4	....	< 1,450 abs. continuous
Methyl iodide.....	CH <sub>3</sub> I	....	10.12	....	3,600-2,110 abs. continuous 2,100-1,215 abs. diffuse 736 emission
Neon.....	Ne	16.6	21.5	16.5	....
Nitric oxide.....	NO	....	9.5	....	....
Nitrogen.....	N <sub>2</sub>	6.1	15.5	6.27	....
Nitrogen dioxide...	NO <sub>2</sub>	....	11.0	....	5,700-2,200 abs. bands
Nitrous oxide.....	N <sub>2</sub> O	....	12.9	....	3,000-1,760 abs. continuous 1,520-1,056 abs. bands < 1,000 abs. continuous
Oxygen.....	O <sub>2</sub>	6.	12.5	3, 5, 9	....
Pyridine.....	C <sub>5</sub> H <sub>5</sub> N	....	9.8	....	< 2,500 abs. continuous
Sulfur dioxide.....	SO <sub>2</sub>	....	13.1	....	3,800-1,529 abs. bands
Water.....	H <sub>2</sub> O	....	13.0	....	1,240-983 abs. bands
Xenon.....	Xe	8.3	12.1	8.27	1,470 emission

been successfully employed to increase by several times the useful life of very small counters [8]. By attaching a bulb of larger dimension to the counter outside the sensitive region to serve as a reservoir, the percentage decomposition of quenching gas per discharge can be made very much smaller.

**10.7. Low-voltage Counter Tubes.** A considerable reduction in the Geiger-Müller threshold of counter tubes containing conventional gas mixtures can be achieved by reducing the total gas pressure. The minimum threshold voltage of most counters containing permanent gases is found at pressures near a few centimeters of mercury, below this the threshold again increases very rapidly with decreasing pressure. With a quenching gas present, the minimum threshold for counters of moderate size is near 500 volts and without a quenching gas, somewhat lower. In practice, however, a substantial reduction in the threshold by this means is impracticable since the reduced pressure is accompanied by a marked decrease in the counter-tube efficiency and ultimately, below 5 to 10 cm Hg, by the alteration or even loss of the Geiger-Müller plateau. It is well known that the threshold voltage also decreases with the diameter of the cathode and to some extent with the diameter of the anode wire, but the reduction effected by the use of very small anode-wire diameters is not very great and is severely limited for mechanical reasons.

Geiger-Müller and proportional counters with very low thresholds can be made, however, with highly specific mixtures of permanent gases and suitable cathode surfaces. Such counters, with operating voltages of 130 to 250 volts and normal characteristics, have been developed and exhaustively investigated by Simpson [30].

The discharge mechanism of these counters is similar to that described in Secs. 10.1 and 10.2 for ordinary gas mixtures but with one important difference. In conventional gas mixtures or with a single permanent gas, a considerable fraction of the energy derived from the electric field during a discharge is taken up by atoms that are raised to metastable excited states rather than being ionized. The energy absorbed by these atoms therefore is largely lost since they do not contribute to the total ionization in the discharge. With the admixture of a small quantity of a second permanent gas, the metastable states can be rapidly reduced by collisions of the second kind, *i.e.*, by inelastic collisions in which a neutral secondary gas atom is ionized and the metastable atom reduced to the ground state [30,31]; thus, more efficient utilization of the electric field for the formation of ions is possible and the threshold voltage is accordingly reduced.

The mechanism described above is possible when the first ionization potential of the secondary gas is less than but near the energy of the metastable state of the primary component. In addition, the concentration of the

secondary gas should be considerably less than 1 per cent of the gas mixture. Combinations of permanent gases suggested by Simpson are given in Table 34.

TABLE 34. LOW-VOLTAGE COUNTER FILLING MIXTURES

Primary component		Secondary component	
Gas	Metastable state, ev	Gas	Ionization potential, ev
Neon.....	16.6	Argon.....	15.7
Helium.....	20.	Neon.....	21.47
Neon.....	16.6	Mercury.....	10.39

Under most conditions, the minimum threshold voltage is found for primary gas pressures of 5 to 10 cm Hg. The optimum amount of secondary gas depends to some extent on the total pressure and kinds of gases used. For the neon-argon mixture, the most carefully investigated, the minimum threshold of approximately 120 volts is found for a neon pressure of  $\sim 5$  cm Hg and  $\sim 0.01$  per cent argon. With argon concentrations up to 0.9 per cent the threshold under the same conditions increases to approximately 160 volts.

Construction, evacuation, and purity of the gases require particular consideration. The cathode surface should be one that minimizes secondary electron emission from both photoemission and ion bombardment. For this reason a thin coating of cuprous oxide gave the most satisfactory results. Before filling, a counter is thoroughly outgassed by pumping for 4 to 6 hr while its temperature is maintained by a furnace at  $300^{\circ}\text{C}$ . Complete outgassing is further ensured by heating the anode wire to incandescence and by applying high-voltage discharges between anode and cathode.

The addition of an organic vapor to make the counter self-quenching raises the threshold to 230 volts or more, but this is still low compared with that of conventional gas mixtures which have thresholds of 800 to 1,500 volts. Specifications for a self-quenching low-voltage counter developed by Simpson [30] are given below.

Cathode: Copper, 1.9 cm diameter

Anode: 0.01 cm diameter tungsten wire

Filling gas: Neon, 5.7 cm Hg; argon 5 to  $50 \times 10^{-4}$  cm Hg; distilled ethyl alcohol, ethyl ether, or amyl acetate, 0.02 to 0.04 cm Hg

Threshold: 230 to 270 volts

Plateau: 90 to 120 volts

Slope: 2 to 5 per cent

**10.8. Active Gas-filled Counters.** A higher order of sensitivity as compared with the thin window counter can be attained in measuring the activity of very low-energy beta-emitting substances by introducing the isotope in some gaseous form into the counter as part of the filling gas. This is particularly useful for measurements of weak activities of  $C^{14}$  (154 kev) and for all activities of  $H^3$  (17 kev). In principle, any radioactive isotope that can be put into gaseous form can be measured by this means. If, however, the maximum energy is greater than 0.2 mev or if gamma rays are emitted, a procedure using a thin-window beta counter or a gamma counter is in general faster and more convenient.

Beta emitters with maximum energies as low as tritium (17 kev) necessarily must be measured under conditions that avoid any absorber including self-absorption. This is possible with counter tubes only by introducing tritium directly into the sensitive region of the tube. Satisfactory procedures of this kind have been developed by Black and Taylor [10], Pace [11], Allen [12], and Cornog [13]. The tritium is introduced as part of the gas filling mixture in the form of HTO vapor. A conventional design of glass-walled tube is used which has a total volume of 1 liter, a 5-cm-diameter copper-screen cathode, and a 0.010-in.-diameter tungsten anode.

The disturbing effects of water vapor in counter tubes are avoided by limiting the HTO vapor pressure in the tube to 2 mm Hg. To this is added a mixture of 2.5 cm Hg of anhydrous ethyl alcohol and 2 cm Hg of argon. A plateau of 300 volts and a threshold of approximately 1,200 volts are reported for this tube by Pace [11].

Memory effects due to absorption of water vapor are considerably more serious than for most other substances, but a procedure suggested by Pace is successful in reducing the counting rate to normal background after each sample measurement. Between samples the tube is alternately evacuated and flushed eight times with inactive water vapor and then followed by an air rinse and finally evacuation to a pressure of 0.3  $\mu$ .

A successful type of gas counter for measuring  $C^{14}$  in the form of carbon dioxide has been developed by Miller [14]. A conventional gamma counter tube is constructed of either metal or glass with a 0.006-in.-diameter tungsten anode and, in the glass tubes, a chemically deposited silver cathode coated with aqua dag. Carbon dioxide containing the  $C^{14}$  is introduced at pressures from 10 to 50 cm Hg. Satisfactory Geiger-Müller characteristics are obtained by the addition of 2 cm Hg of carbon disulfide which provides a 200-volt plateau with a 2 per cent slope. The threshold voltage varies from 1,800 to 4,500 depending upon the cathode diameter (1 to 4 cm) and gas pressure. The optimum operating voltage was found to be approximately 160 volts above threshold voltage.

Memory effect appears to be negligible for carbon dioxide.

**10.9. Neutron Counters.** Counters intended for direct detection of neutrons depend upon either the recoil of light nuclei in the filling gas and from the walls, or on charged particles emitted in nuclear interactions of neutrons with the material of the counter wall or filling gas. The choice between these two mechanisms depends upon the energy range of the neutrons to be detected. Whichever method is used, in most applications neutron counters are operated in the proportional region in order to reduce the less heavily ionizing background of gamma and electron radiation. This is essential for measuring low neutron flux since the efficiency for neutrons is small compared with that for incident charged particles.

**10.10. Slow Neutron Counters.** Slow neutrons are usually detected by their interaction with  $B^{10}$  nuclei according to the reaction  $B^{10}(n, \alpha)Li^7$ . The reaction is accompanied by the release of 2.5 mev of which 1.6 mev is contributed to the alpha particle and 0.9 mev to the recoil lithium nucleus. If both particles are stopped in the filling gas, roughly 75,000 ion pairs are formed, assuming an average energy loss of 33 ev per ion pair. Boron can be used in slow neutron counters either as the filling gas in the form of boron trifluoride [15] or as a thin coating of metal on the cathode wall.

Boron trifluoride-filled counters are constructed similarly to gamma counters, preferably with materials that do not have large thermal-neutron capture cross sections, as does glass containing boron for example, and filled to pressures of 10 to 760 mm Hg. The maximum pressure that can be used is limited by both the increase in operating voltage and the increased size of electron pulses as the boron trifluoride pressure is raised. The lowest useful pressure is determined by the neutron counting efficiency desired.

The efficiency of the counter, defined as the probability that a neutron in traversing an average path length  $l$  through the counter is captured, is given [6] by,

$$\epsilon = lrpL\sigma$$

where  $p$  = pressure

$L$  = Loschmidt's number

$r$  =  $B^{10}$  concentration,  $B^{10}/(B^{10} + B^{11})$

$\sigma$  =  $B^{10}$  capture cross section for neutrons of energy  $E$

The neutron capture cross section of  $B^{11}$  is negligible compared with  $B^{10}$  and does not contribute appreciably to the neutron count. Higher counting efficiency therefore can be attained by increasing the enhancement of  $B^{10}$  above its natural fractional concentration of 0.18 [16]. The efficiency also varies with the neutron energy since for boron  $\sigma \sim E^{-1/2}$ . Within the experimental accuracy, the cross section for energies between 0.01 and 10,000 ev is given by

$$\sigma = \frac{114}{\sqrt{E}} - 0.20 \quad \text{barns}$$

where  $E$  = neutron energy, ev



Neutron counters with boron-coated cathodes are usually filled with conventional argon-alcohol mixtures and operated at voltages within the proportional region for the particular counter. The thickness of the boron layer on the cathode should not be made greater than the range of the alpha particle ejected from a boron nucleus by a slow neutron capture, *i.e.*, about 0.1 mm. Greater thicknesses will not increase the efficiency but, rather, lead to excessive absorption of neutrons since a larger fraction of ejected alpha particles will not reach the sensitive volume of the counter. The maximum efficiency of such counters is given [6] by the formula

$$\epsilon = \frac{\rho \sigma R N}{A}$$

where  $\rho$  = boron density

$N$  = Avogadro's number

$A$  = atomic weight

$R$  = alpha-particle range in boron

$\sigma$  = boron cross section for neutrons of energy  $E$

In a similar way, a very thin layer of uranium may be used instead of boron. The fission products resulting from the absorption of slow neutrons produce extremely heavy ionization. However if most of the range of a fragment lies within the uranium layer, it cannot be distinguished from the alpha particles due to the natural uranium decay and must, therefore, be treated by the usual statistical methods for background counts. The normal fractional concentration of the effective isotope is 0.007 and has a cross section  $\sim 550$  barns for 0.025 ev neutrons. The use of enhanced  $U^{235}$  mixtures will increase the efficiency correspondingly.

**10.11. Fast Neutron Counters.** Detection of fast neutrons with counters is accomplished most effectively by the recoil of light nuclei from elastic collisions with neutrons. The maximum energy transferred from the neutron to the struck nucleus is

$$E_r = E \frac{4M}{(M+1)^2} = E\alpha \quad \text{mev}$$

where  $M$  = mass of nucleus in units of neutron mass

$E$  = neutron energy

$E_r$  = recoil kinetic energy of nucleus

and the average energy is approximately

$$\bar{E} = E \frac{2M}{(M+1)^2}$$

If hydrogen is used for the filling gas, the maximum recoil proton energy, due to a head-on collision, is equal to the full neutron energy.

If, for a gas of atomic mass  $M$ , the smallest detectable pulse is produced by a recoil energy  $B_0$ , referred to as the bias energy, the lowest neutron energy

that can be detected is  $B = B_0 \alpha$ . For hydrogen,  $B = B_0$ , and the scattering cross section for neutron energies above 0.050 mev varies as  $1/E^{1/2}$ . Therefore, the sensitivity of hydrogen-filled counters varies approximately as [17]

$$E^{-1/2} \left( 1 - \frac{B_0}{E} \right)$$

The sensitivity rises rapidly for neutrons with greater than the bias energy and then remains relatively constant in the energy interval

$$1.57 B < E < 9.6B.$$

The existence of a threshold is an important property of the hydrogen-filled proportional counter since it provides a rapid although rough means for determining neutron energies. This is done by altering the bias of a discriminator to pass only pulses due to recoil nuclei with energies equal to or greater than  $B_0$ .

An alternative form of proton-recoil counter employs thin hydrogenous radiators mounted within the counter tube [17,18]. Radiators are frequently prepared by evaporation of glycerol tristearate on thin platinum disks in a vacuum. The thickness of the hydrogenous layer is usually of the order of 100 mg per cm<sup>2</sup>. The same characteristics are observed as for hydrogen-filled counters provided that the filling gas has a sufficiently high stopping power to stop all hydrogen recoils within the gas volume and does not itself recoil with greater than the bias energy  $B_0$ . Heavy, inert gases such as argon, krypton, and xenon are the most satisfactory filling gases for this reason.

The elastic scattering cross section of hydrogen and most other substances for fast neutrons is in the order of 1 barn; hence the efficiency of counters used in this energy range is necessarily low. In addition to counts from recoil nuclei, counts will be registered from competing neutron processes occurring in the counter wall and in the filling gases, liberating protons, deuterons, alpha particles, or gamma rays. The cross sections for these interactions in nearly all substances is also in the order of 1 barn. Thus, if a boron trifluoride counter is used for detecting very fast neutrons, discharges are initiated by alpha particles from the reaction  $B(n, \alpha)Li$  as well as by recoil boron and fluorine nuclei.

**10.12. Accuracy of Counting Measurements.** An estimate of the accuracy of counting measurements on the activity of a sample is determined, for the most part, by simple statistical procedures. The methods employed are based upon the fact that each disintegration is a statistically independent event since it is in no way affected by a preceding event or the means by which it is detected. The distribution of disintegrations in time and the

radiation detected is then purely random. In a time interval  $t$ , which is small compared to the half-life of the substance, the probability  $P$  of observing  $n$  particles is given [19] by Poisson's distribution curve for random events.

$$P(n) = \frac{N^n}{n!} e^{-N}$$

where  $N$  is the true average number of particles detected per unit time. For large numbers of counts the difference between the true average and the computed average (the most probable value) is always small, and no significant error is introduced by using the latter value.

Random errors accompany all measurements; they are indeterminate and presumably arise from a multitude of unknown factors which influence slightly the measured values. They can, however, be treated by statistical methods which provide a measure of the possible error introduced by random fluctuations. The true value of a quantity, in this instance the counting rate, is seldom known, but in its place the most probable or average value of the set of measurements must be used. Residuals may now be defined as the difference between each measurement  $n_i$  and the average value  $\bar{n}$ ;  $x_i = n_i - \bar{n}$ . If the residuals are truly random, there will be about the same number of negative as positive values, and small values will occur with greater frequency than large values. It can be shown that under this condition the distribution of the residuals is nearly symmetrical about the average value and follows approximately the Gauss error curve (normal distribution curve)

$$P(x) = \frac{e^{-x^2/2\sigma^2}}{\sigma \sqrt{2\pi}} = \frac{h}{\sqrt{\pi}} e^{-hx^2}$$

where  $\sigma$  = standard deviation (see below)

$$h = 1/2\sigma^2 = \text{index of precision}$$

If  $h$  is large, a greater proportion of the errors are small and hence grouped close to  $\bar{n}$ ; if  $h$  is small, the spread in data is greater.

Aside from the intrinsic form of the distribution of data and errors to be expected, the important facts concerning a set of measurements are estimates of reliability in terms of the variability of the data. Three measures of variability are commonly used for this purpose: average deviation from the mean, standard deviation from the mean, and the probable error. Of these, the probable error is most often used for estimating the statistical error in counting measurements.

**10.13. Average Deviation.** If  $\bar{n}$  is the average number of counts per unit time for  $M$  measurements of the same sample under identical physical conditions, the average deviation of the residuals from the mean value of the set of measurements is (in counts per minute)

$$A.D. = \frac{\sum_{i=1}^M |\bar{n} - n_i|}{\sqrt{M(M-1)}} = \frac{1}{h\sqrt{\pi}} \quad \text{cpm}$$

If  $M \gg 1$ ,

$$A.D. = \frac{\sum_{i=1}^M |x_i|}{M} \quad \text{cpm}$$

where

$$x_i = n_i - \bar{n}$$

$$\bar{n} = \frac{\sum_{i=1}^M n_i}{M} \quad \text{cpm}$$

and the vertical bars indicate absolute values. These formulas may be applied when counting by predetermined time intervals  $t_i$ , or by predetermined count number intervals  $n_i$ , or by arbitrary choice of both.

**10.14. Standard Deviation.** The standard deviation  $\sigma$  of a measurement is defined as the root mean square of the deviations from the mean value of the counting rate.

$$\sigma = \sqrt{\frac{\sum_{i=1}^M |\bar{n} - n_i|^2}{M-1}} \quad \text{cpm}$$

Also a more convenient form is generally used when a single measurement of  $N$  total counts is taken over a time  $t$

$$\sigma = \sqrt{\bar{n}t} = \sqrt{N} \quad \text{counts}$$

which is the standard error in terms of numbers of counts. In terms of percentage,

$$\sigma = \frac{100}{\sqrt{N}} \quad \%$$

If several sets of measurements are taken, the resulting standard deviation is then the square root of the sum of the squares of the separate  $\sigma$ 's

$$\sigma^2 = \sum_{i=1}^M \sigma_i^2$$

Either this method or the one above may be used for computing  $\sigma$ . The error in any counting measurement must necessarily include the standard

deviation of the background count  $\sigma_b$  as well as of the sample plus background  $\sigma_s$  and is given by

$$\sigma = \sqrt{\sigma_b^2 + \sigma_s^2} = \sqrt{N_b + N_s} \quad \text{counts}$$

$N_b$  and  $N_s$  are the number of registered counts for background and the sample plus background taken over equal intervals of time.

**10.15. Probable Error.** The probable error  $r$  is calculated from the standard deviation  $\sigma$  or directly from the total number of counts of the sample plus background,  $N_s$ , and the total background count  $N_b$  counted for the same length of time. The probable error in number of counts is

$$r = 0.6745\sigma = 0.6745 \sqrt{N_s + N_b} \quad \text{counts}$$

In terms of per cent,

$$r = \frac{67.45}{(N_s + N_b)} \sqrt{N_s + N_b} \quad \%$$

It is seen that, like the standard deviation, the probable error may be calculated from the total number of counts  $N$  or from the  $M$  sets of measurements.

The probable error defines limits,  $\pm r$ , about the mean value,  $\bar{N}$  counts in time  $t$ , or  $\bar{n}$  cpm, within which any single measurement of the activity should occur with a probability of 0.5 and with an equal probability for all values outside these limits.

When the approximate values of the sample and background counting rates are known, it is frequently more convenient to count samples by time intervals rather than by total events. When the sample plus background and the background are counted separately to give the same relative probable error, the minimum time required to reduce the total statistical error of the sample measurement to a prescribed percentage  $r$  is given by

$$t = \frac{9,100\bar{n}}{r^2(\bar{n} - \bar{n}_b)^2} \quad \text{min}$$

and for the background, the minimum counting time should be

$$t_b = \frac{(\bar{n} - \bar{n}_b)^2 t}{\bar{n}\bar{n}_b} \quad \text{min}$$

where  $\bar{n}$  = average counting rate of sample plus background

$\bar{n}_b$  = average counting rate of background

The true value of  $\bar{n} - \bar{n}_b$  is then calculated with the prescribed accuracy from the total counts  $N$  in the time  $t$  and background counts  $N_b$  in the time  $t_b$ .

When possible, lead shielding should be used to reduce the magnitude of the background counting rate and hence the required counting time.

**10.16. Counter Resolving Time.** The shortest interval in which two successive events can produce in a counter separate discharges of sufficient amplitude to register as counts is referred to as the resolving time  $\tau$ . Since the actual insensitive time of a counter following a discharge varies somewhat, the measured value of  $\tau$  is the average of these intervals. For most counters the resolving time is in the order of  $10^{-4}$  sec, which is approximately the collection time of the positive ions formed in the discharge.

The value of  $\tau$  for any counter can be ascertained by two general methods in use at the present time. The first is an indirect method in which  $\tau$  is calculated from the observed counting loss, and the second is a direct method where the length of the interval is measured with electronic devices such as the synchroscope.

The first of these methods is most often used since it requires no special equipment and can be carried out relatively quickly. Two samples, I and II, with nearly equal activities are prepared and measured as follows: Sample I is counted alone giving a counting rate  $n_1$ ; without disturbing it sample II is placed in counting position and the total counting rate  $n_{12}$  of I + II observed; sample I is then removed and the counting rate  $n_2$  of sample II obtained. From the three counting rates and that of the background  $n_b$ , the resolving time is calculated [35] with the formula

$$\tau = \frac{n_1 + n_2 - n_{12} - n_b}{n_{12}^2 - n_1^2 - n_2^2} \quad \text{min}$$

The total number of counts in each measurement should be sufficiently large and of roughly the same magnitude to ensure a small probable error in the counting rate since  $\tau$  is estimated from small differences between large numbers.

The formula given here is one of several that have been suggested for computing the resolving time from a single set of measurements with a paired source. Although the various formulas differ considerably at high counting rates, they lead to nearly the same value for  $\tau$  at low rates where the formula above is valid. It is essential therefore to use test samples whose activities provide counting rates at levels where  $100 (n_1 + n_2 - n_{12})/n_{12}$  is no greater than several per cent. At the same time, however, the activities should be high compared to the background count to ensure accuracy in a reasonable length of time. Normally, samples with counting rates in the order of 1,000 cpm are found most useful.

The direct method for evaluating  $\tau$  requires a linear pulse amplifier and an oscilloscope with accurately known sweep frequencies. The width of the pulse in microseconds can be estimated directly from the oscilloscope trace and taken as a first approximation for  $\tau$ . A more useful device is an oscilloscope equipped with a single sweep circuit and a persistent screen. The

horizontal sweep is triggered by incoming pulses which are then traced at a fixed position on the screen and allow more careful measurement. This provides a better opportunity to observe closely spaced pulses and hence to verify the resolving time.

**10.17. Coincidence Corrections for High Counting Rates.** Pulse-counting devices such as counter tubes, ionization chambers, and electrical circuits are never linear in their recording rate when the events registered occur in random intervals. As the average counting rate increases, a greater proportion of the events producing the counts occurs in intervals shorter than the resolving time of the device, and therefore pairs and triplets of events are more often registered as a single count. This increase in coincidence rate of multiple events causes the response of a counter to deviate markedly from a linear relation to source activity. Unless a coincidence correction is made, the observed counting rates at high levels ( $> 10,000$  cpm) have little significance and even at levels greater than 1,000 cpm may be in serious error. The coincidence correction therefore must determine from the observed counting rate the true rate of events or that rate which would be observed if the resolving time were zero.

It is apparent that the true counting rate should be a function of the resolving time and the registered rate, but it also depends on the mechanism of recovery since two distinct alternatives present themselves. The first mechanism leads to a correction formula [35] in the form

$$N = n + \tau n^2 \quad \text{cpm}$$

where  $N$  = true counting rate

$n$  = recorded counting rate

$\tau$  = counter resolving time

At low counting rates where  $N \simeq n$ , the more convenient approximation

$$N = n + \tau n^2 \quad \text{cpm}$$

can be used. This formula and the preceding one are derived on the assumption that the insensitive time is more or less independent of events that occur immediately before or after a count is recorded and, in particular, the occurrence of an unrecorded event during recovery of the device does not extend the insensitive interval. Although this is not strictly true for counters, the influence of this factor is small and the formulas above are the most nearly valid in this case. It is seen from the formula that as the true activity increases to very high levels the recorded counting rate approaches asymptotically the limiting value  $1/\tau$  as  $N \rightarrow \infty$ ; counts are recorded only as fast as the tube can recover.

The second correction formula applies less to counters but represents more accurately the counting loss in electronic and mechanical devices. It

is given by Volz [41] and Schiff [39] as

$$n = Ne^{-\tau n} \quad \text{cpm}$$

It is derived on the assumption that the occurrence of an event while the device is recovering from previous count is not counted but has the effect of prolonging the recovery as though it had been counted. It is evident from this that as the true activity increases without limit the recorded counting rate goes to zero; the device becomes blocked and no counts are passed. At low counting rates, however, the difference between this and the preceding formula is negligible and either one can be validly used.

It is tacitly assumed in applying these formulas that the resolving time of the counter tube is longer than that of all components of the circuits which follow, including that of the register divided by the scaling factor. If this were not true, the formulas above would not be valid since the resolving time of such components would also influence the amount of the correction. Fortunately this requirement is met in modern counting circuits, for the exact calculations in this case prove to be impracticable.

Nevertheless, at very high counting rates the correction formulas above are not sufficiently accurate to give reliable values for the true counting rate. Consequently it is often necessary to construct a calibration curve with the requisite accuracy extending to very high counting-rate levels. This can be done analytically with any prescribed accuracy by expressing the true counting rate in terms of a power series in  $n$ , the recorded rate, of the form

$$N = n + \tau n^2 + \nu n^3 + \mu n^4 + \quad \text{cpm}$$

An arbitrary number of terms may be used, depending upon the desired accuracy, but the labor involved in computing the coefficients of terms beyond the fourth usually makes the inclusion of higher order terms impracticable. The coefficient of the first term is unity because at very low counting rates  $N \simeq n$ . The second coefficient is the counter resolving time. The physical significance of higher order coefficients is less clear other than that they are coefficients of higher order moments which take into account variations from the simple quadratic curve due to second-order effects that influence the counting rate.

Kohman [33] has presented in detail a method for evaluating the coefficients from a least-squares solution of measurements on paired sources. Each set of measurements includes the recorded counting rates  $n_1$  of source I,  $n_2$  of source II, which is approximately equal in activity to I, their combined rate  $n_{12}$ , and the background count  $n_b$  (see preceding section for experimental procedure). For the greatest accuracy, many sets of measurements should be made with paired sources covering the entire counting range.



With data thus obtained the coefficients are calculated which make the quantity

$$\frac{(N_1 + N_2 - N_{12} - n_b)^2}{n_{12}^2}$$

a minimum. This is simply an expression of the principle of least squares stating that the curve which best fits the data is that for which the sum of the squares of the differences between the observed points and the curve is a minimum.

When only two terms of the series are used, the single coefficient is calculated [33] by the formula

$$= \frac{\sum \frac{1}{n_{12}} (n_1 + n_2 - n_{12} - n_b)(n_1^2 + n_2^2 - n_{12}^2)}{\sum \frac{1}{n_{12}} (n_1^2 + n_2^2 - n_{12}^2)^2} \quad \min$$

in which the sum is taken over the  $s$  sets of measurements of  $n_1$ ,  $n_2$ , and  $n_{12}$

When three terms of the series are desired, the two coefficients  $\tau$  and  $\nu$  are calculated from the  $s$  sets of data [33] by the formulas

$$\begin{aligned} \tau &= \frac{JK - HM}{LM - K^2} & \min \\ \nu &= \frac{HK - JL}{LM - K^2} & \min^2 \end{aligned}$$

where

$$\begin{aligned} H &= \sum \frac{1}{n_{12}^2} (n_1 + n_2 - n_{12} - n_b)(n_1^2 + n_2^2 - n_{12}^2) \\ J &= \sum \frac{1}{n_{12}^2} (n_1 + n_2 - n_{12} - n_b)(n_1^3 + n_2^3 - n_{12}^3) \\ K &= \sum \frac{1}{n_{12}^2} (n_1^2 + n_2^2 - n_{12}^2)(n_1^3 + n_2^3 - n_{12}^3) \\ L &= \sum \frac{1}{n_{12}^2} (n_1^2 + n_2^2 - n_{12}^2)^2 \\ M &= \sum \frac{1}{n_{12}^2} (n_1^3 + n_2^3 - n_{12}^3)^2 \end{aligned}$$

**10.18. Coincidence Counting Corrections.** As is done for single counters, coincidence counting measurements must be corrected for background, in

this case only for accidental coincidence counts. Such counts may be produced by the simultaneous arrival of several related particles as in cosmic-ray showers or by the chance arrival of two independent particles in a time of the order  $2\tau$  due to a high radiation level such as exists near high-energy accelerators. The number of accidental counts per minute to be expected from a twofold coincidence arrangement is

$$m = 2\bar{n}_1\bar{n}_2\tau \quad \text{cpm}$$

where  $\bar{n}_1, \bar{n}_2$  = average counts per minute independently from counters 1 and 2

$\tau$  = resolving time of counters, min

For an  $r$ -fold coincidence arrangement, assuming the resolving times to be equal, the accidental counting rate is

$$m = r\tau^{r-1}(\bar{n}_1\bar{n}_2 \cdots \bar{n}_r) \quad \text{cpm}$$

If the resolving time of each of the counters in an  $r$ -fold coincidence set is different, the accidental counting rate is [22]

$$m = \bar{n}_1\bar{n}_2 \cdots \bar{n}_r\tau_1\tau_2 \cdots \tau_r \left( \frac{1}{\tau_1} + \frac{1}{\tau_2} + \cdots + \frac{1}{\tau_r} \right) \quad \text{cpm}$$

If the radiation density is not high, the accidental rate can be determined by dispersing the counters and counting the coincidences. Alternatively, the accidental rate can be made negligibly small by increasing the multiplicity of the system and by reducing the counter resolving time. Accidentals may be further reduced by the proper use of anticoincidence counters; however, their use decreases the counting efficiency because of the over-all increase in resolving time of the set.

**10.19. Counter Efficiency.** Aside from the counting loss due to the finite resolving time, not all particles traversing a counter will produce discharges. The efficiency with which a counter responds to incident particles can be determined with a three- or higher fold coincident arrangement by counting coincidences with and without the counter as a part of the system. The absolute counter efficiency is then given by

$$\epsilon = \frac{n_r - m_r}{n_{r-1} - m_{r-1}}$$

where  $n_r$  = total counts in time  $t$  (or counting rate) of  $r$ -fold coincidence arrangement, *i.e.*, with tested counter included

$n_{r-1}$  = same but with tested counter removed

$m_r$  = accidental counts in time  $t$  (or counting rate) for  $r$ -fold coincidence arrangement, *i.e.*, with tested counter included

$m_{r-1}$  = accidental counts without tested counter

With the efficiency of each counter known, the total efficiency of an  $r$ -fold coincidence arrangement is simply the product of the separate efficiencies or  $\epsilon_t = \epsilon_1 \epsilon_2 \cdots \epsilon_r$ .

**10.20. Averaging Effect of Scaling Circuit.** The slowest component in a counting circuit is the mechanical register. Even the most efficient registers are slower than the counter tube by several orders of magnitude. In most cases, therefore, scaling circuits are advisable even at moderate counting rates. For a scale factor  $s$  and a register resolving time  $T_1$  no counts are lost or corrections necessary if  $T_1 \leq s\tau$ , where  $\tau$  is the counter-tube resolving time, since the counter tube cannot produce two pulses in an interval shorter than the resolving time of the register. On the other hand, it more frequently occurs that  $T_1 > s\tau$ , indicating that pairs of output pulses from the scaling circuit can occur in intervals shorter than the register can function. A scaling circuit, however, tends to average the intervals between output pulses since it passes only one pulse after receiving  $s$  random input pulses and the intervals tend to be more uniform. For high scale factors, 32 and greater, the probability that pulse intervals for a counting rate  $n$  deviate markedly from  $s/n$  is small. Thus, for a scale of 64, the standard deviation of the lengths of intervals from the average is only one-eighth the average interval [36]. For most purposes, register losses are negligible when  $T_1 < s/n$  for scale factors of 32 and greater. At high counting rates, however, it is usually desirable and often necessary to use scale factors of 128 or decade circuits of 1,000.

**10.21. Quenching Circuits.** Quenching circuits serve to extinguish the discharge in a counter tube by suddenly lowering the tube potential below the voltage at which a discharge can continue. This action should occur in an interval that is short as compared to the drift time of the positive ions, and the operating voltage should be returned to the tube by the time all positive ions have been swept out. The recovery time of the external circuit should be shorter than that of the counting tube but not so short as to allow continuation of the discharge. Generally it is in the order of  $10^{-8}$  sec.

External quenching circuits are essential to the operation of non-self-quenching counter tubes but usually are not necessary for self-quenching counters. Nevertheless, they are frequently used as the first stage for any

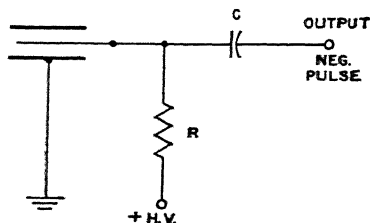


FIG. 79. Resistance quenching circuit. When the counter tube discharges the current that flows through the tube is sufficient to lower the potential across  $R$  below the Geiger-Müller threshold voltage. The time constant of  $10^{-2} - 10^{-3}$  sec is too long for most applications.  $R = 10^7 - 10^9$  ohms,  $C = 2 - 10 \mu\text{mf}$ .

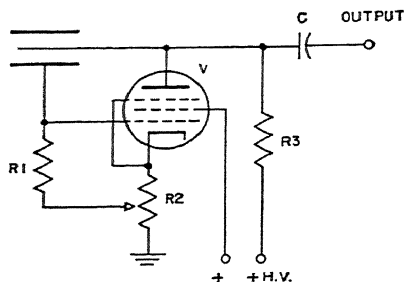


FIG. 80. Neher-Harper quenching circuit. The grid bias voltage is adjusted at  $R_2$  so that tube  $V$  is normally nonconducting. When the counter tube discharges, the grid is driven positive and the tube conducts, thus producing the necessary voltage drop in resistance  $R_3$  to stop the counter discharge. The time constant of the circuit is determined by  $R_3$  and  $C$ . Approximate values of the constants are  $R_1 = 10^6 - 10^7$  ohms,  $R_3 = 10,000$  ohms and  $C = 100 \mu\text{fd}$ . [C. E. Wynn-Williams, *Brit. Pat.*, 421 341 (1934); H. V. Neher and W. W. Harper, *Phys. Rev.*, 49, 940 (1936).]

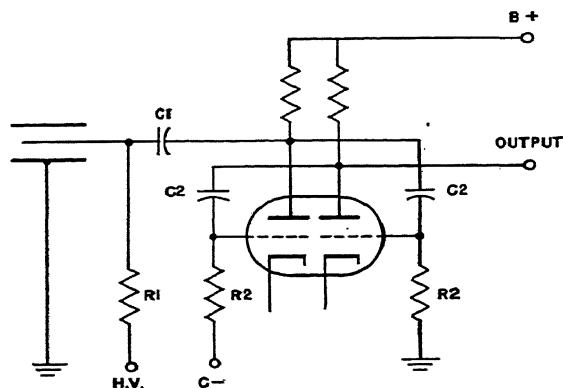


FIG. 81. Multivibrator quenching circuit. The high negative bias,  $-C$  makes first section of the multivibrator nonconducting while the second section is normally conducting. A negative pulse from the counter tube initiates one cycle of multivibrator during which the first section conducts momentarily and thus applies a high negative voltage pulse to the counter tube anode to extinguish the discharge. The time constant of the circuit depends primarily on the values of  $R_2$  and  $C_2$ . [I. A. Gettings, *Phys. Rev.*, 53, 103 (1938).]

kind of counter tube since some quenching circuits also serve as a preamplifier and provide a low-impedance output which permits the use of long connecting cables to the scaling or the recording circuit. Furthermore it is found in many instances that the quenching circuit can, when properly adjusted for the counter, improve the pulse shape and duration, particularly

for old counters. If no quenching or preamplifying circuit is used with a self-quenching counter tube, the lead to the counting circuit should be short and well shielded. Lengths up to several meters usually will not seriously impair the pulse characteristics.

The most frequently used quenching circuits are shown in Figs. 79 to 82.

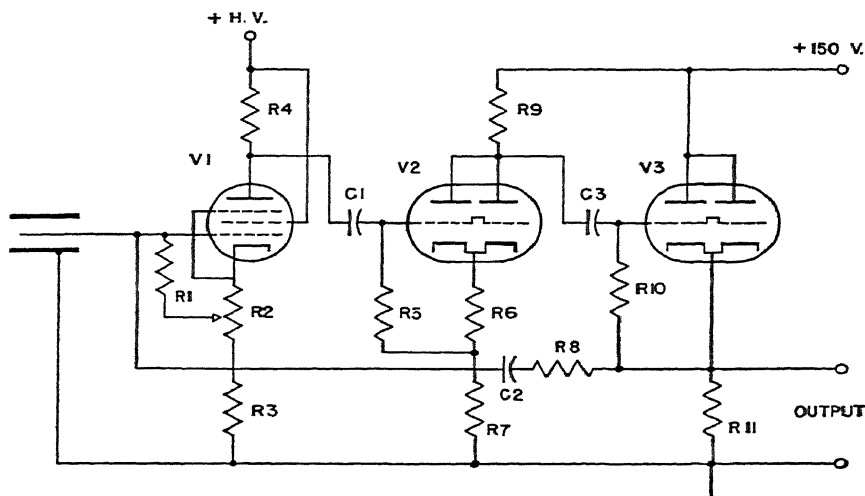


FIG. 82. Modified Neher-Pickering quenching circuit. This circuit allows operation of the counter tube with the cathode grounded. It further provides one stage of amplification (V2), stabilization by negative feedback and a cathode follower output (V3).

$R1, R3 = 1 \text{ megohm}$	$R7, R11 = 20,000 \text{ ohms}$	$C1 = 50 \mu\text{fd}$
$R2, R6 = 10,000 \text{ ohms}$	$R8 = 1.1 \text{ megohms}$	$C2 = 250 \mu\text{fd}$
$R4 = 240,000 \text{ ohms}$	$R9 = 56,000 \text{ ohms}$	$C3 = 100 \mu\text{fd}$
$R5 = 5 \text{ megohms}$	$R10 = 5 \text{ megohms}$	$V1 = 6AK5$
		$V2, V3 = 6J6$

**10.22. Scaling Circuits.** Scaling circuits must be used whenever the counting rate exceeds that which the mechanical register can follow without loss of counts. For the best registers the limiting rate is about 25 to 50 cps. In practice, however, scaling circuits are normally employed for all but the lowest counting rates. Most counter circuits now available are based on the scale of two, with six such scaling circuits in series to provide a total scale factor of 64.

The fundamental scale of two circuit consists of a trigger circuit that has two stable states of operation, the first tube conducting and the second nonconducting, or the reverse. The arrival of a pulse flips the trigger cir-

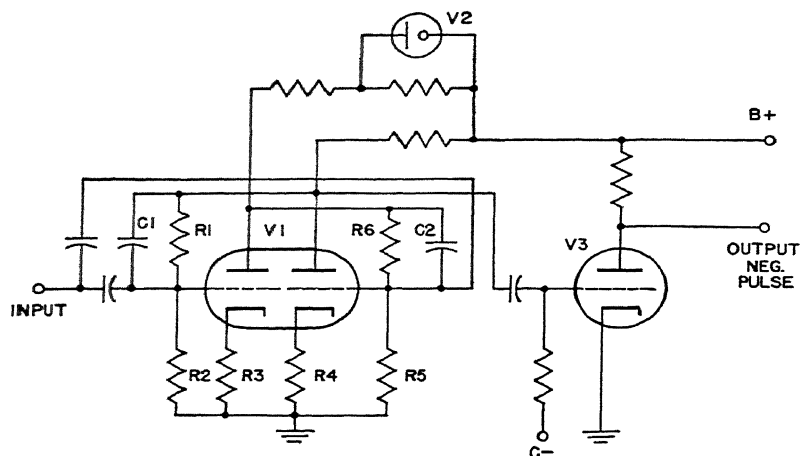


FIG. 83. Scale of two-trigger circuit. The two units of V1 form a trigger circuit delivering alternately a positive and negative pulse. Negative pulses are clipped by V3, thus passing one pulse for every two at the input. Scales of 4, 8, 16, 32, etc., are obtained by adding identical units in series. The neon tube V2 serves as an indicator to extrapolate between multiples of the scale factor. Approximate circuit values are

R1, R6 = 400,000 ohms	V1 = 6N7, 6SL7 or pairs of single triads
R2, R5 = 500,000 ohms	V2 = neon, $\frac{1}{4}$ watt
R3, R4 = 40,000 ohms	V3 = 6C5 or similar triode
C1, C2 = 40 $\mu$ fd	

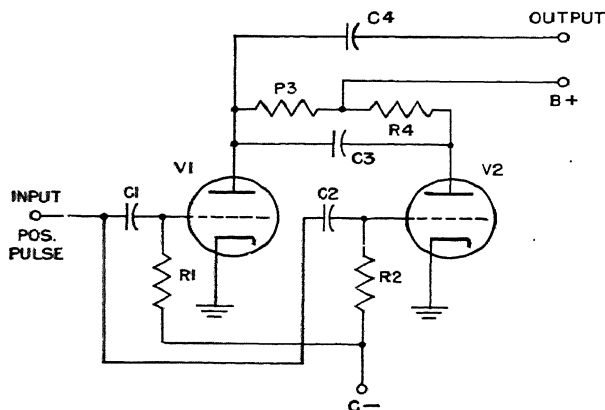


FIG. 84. Thyatron scale of two circuit. Higher scale factors are obtained with additional units in series. Approximate circuit values are R1, R2 = 50,000 ohms; R3, R4 = 20,000 ohms; C1, C2, C3, C4 = 0.002  $\mu$ fd; V1, V2 = Thyatron, e.g., 884.

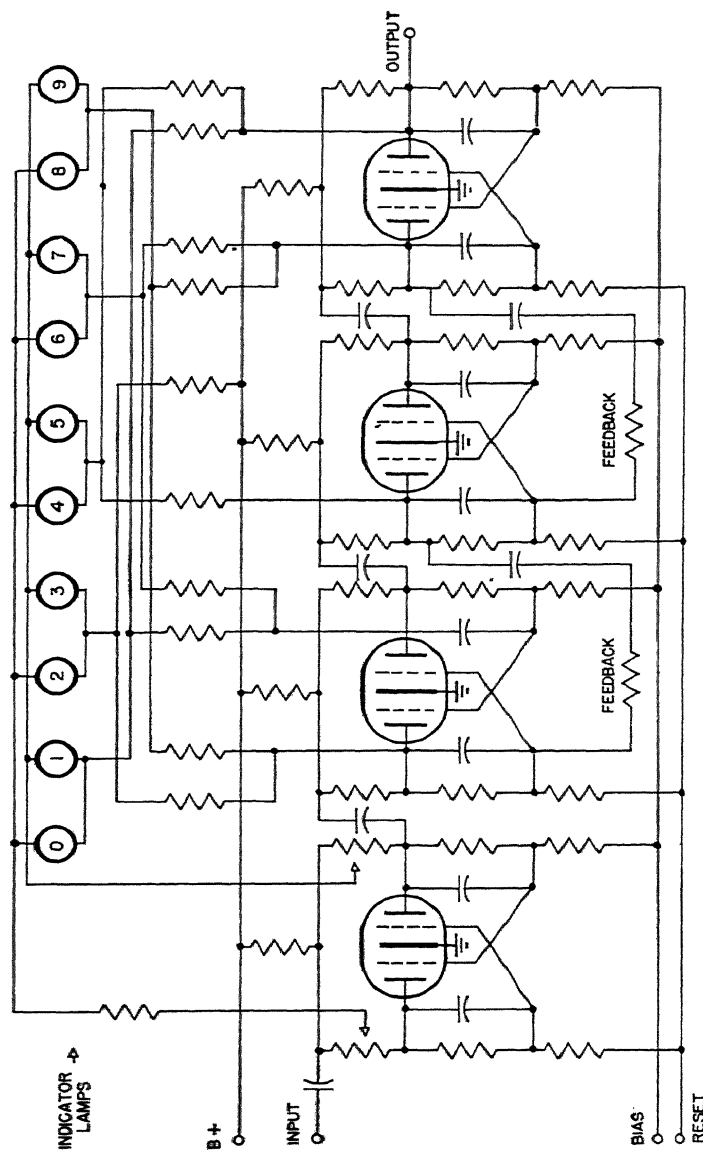


FIG. 85. Binary decade scaling circuit. This decade circuit consists of four resistance-coupled multivibrators with inverse feedback in two stages. The natural count of the circuit is 16, but in the feedback stages the count is advanced each time by the natural count of that stage, thus completing the cycle in 10 counts rather than 16. Only one indicator lamp is lighted for each count from 0 to 9. [*J. E. Grosdorff, R.C.A. Rev., 1, No. 13 (1946).*]

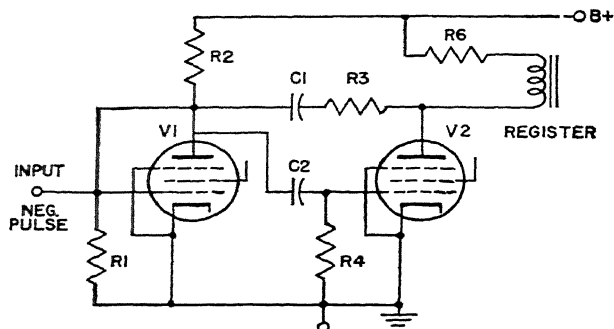


FIG. 86. Multivibrator recording circuit. Approximate values of circuit constants are

$R1 = 1$  megohm  
 $R2 = 100,000$  ohms  
 $R3 = 50,000$  ohms  
 $R4 = 500,000$  ohms  
 $R6 = 5,000$  ohms

$C1 = 0.0001$   $\mu$ fd  
 $C2 = 0.001$   $\mu$ fd  
 $V1 = 6SJ7, 6C6$   
 $V2 = 6V6, 6F6$

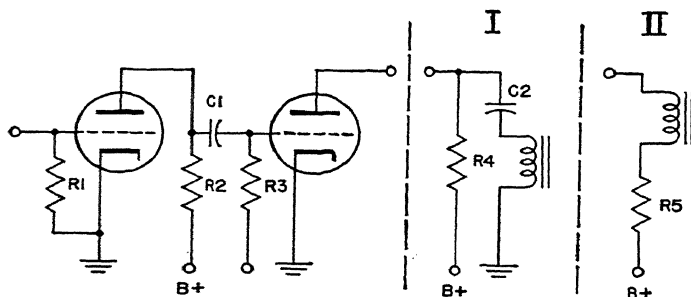


FIG. 87. Thyatron recording circuit. Two arrangements of the register circuit are illustrated to the right of the broken lines. Circuit I is preferred since the recorder is at ground potential and there is less chance of damage to the register due to surges. Approximate circuit values are

$R1, R3 = 500,000$  ohms  
 $R2 = 100,000$  ohms  
 $R4 = 200,000$  ohms  
 $R5 = 10,000$  ohms

$C1 = 0.001$   $\mu$ fd  
 $C2 = 1 - 4$   $\mu$ fd  
 $V1 = 6C5$   
 $V2 = \text{Thyatron; e.g., 884}$

cuit from one state to the other; hence two input pulses of the same sign are required for a complete cycle. Although two output pulses also appear per cycle, they are opposite in polarity and one pulse can be rejected by a "clipping" circuit immediately following each of the trigger circuits. Normally, the negative pulse, which is the first output pulse in the cycle is



rejected. This is usually accomplished either with a multigrid tube with a negative grid bias or with a diode, both of which pass only positive pulses. Typical scale of two circuits are shown in Figs. 83 and 84.

Less frequently used in the past, decade counters and "ring" circuits are now finding more frequent application. An example of a highly efficient decade circuit is shown in Fig. 85.

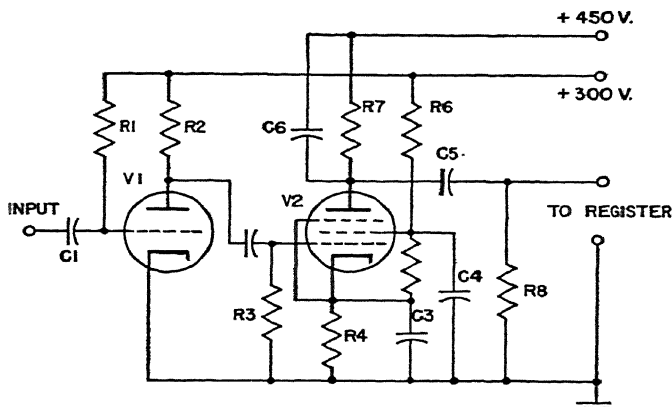


FIG. 88. Recording circuit from "Model 200" pulse counter.  $V1$  reshapes the pulse into a rectangular form before it enters the power tube  $V2$ . The output pulse length is 0.01 sec and delivers sufficient power to operate any register that requires no more than 40 ma and 300 volts. [W. A. Higinbotham, J. Gallagher, and M. Sands, *Rev. Sci. Instruments*, **18**, 706 (1947).]

$R1 = 2$  megohms  
 $R2 = 200,000$  ohms  
 $R3 = 1$  megohm  
 $R4 = 1,500$  ohms  
 $R5 = 10,000$  ohms  
 $R6 = 20,000$  ohms  
 $R7 = 30,000$  ohms  
 $R8 = 100,000$  ohms

$C1 = 0.05$   $\mu$ fd  
 $C2 = 0.005$   $\mu$ fd  
 $C3 = 20$   $\mu$ fd  
 $C4 = 8$   $\mu$ fd  
 $C5 = 4$   $\mu$ fd  
 $C6 = 0.01$   $\mu$ fd  
 $V1 = 6SL7$   
 $V2 = 6V7$

**10.23. Discriminators.** Discriminating circuits are often placed before counter circuits for the purpose of rejecting pulses smaller than a predetermined peak voltage. Their function is particularly useful for proportional counters where the pulse size depends upon the intensity of ionization produced by the incident particle. Thus, in counting alpha particles the background is effectively reduced by rejecting the smaller pulses initiated by electrons, gamma rays, and protons.

With more complicated circuits, pulses lying within a desired voltage range can be passed, whereas pulses smaller and larger are rejected. With several such units, pulses produced by electrons, protons, alpha particles, and fission fragments may be distinguished and recorded separately.

**10.24. Recording Circuits.** The recording or register-driving circuit is essentially a power output stage delivering sufficient power at each pulse to

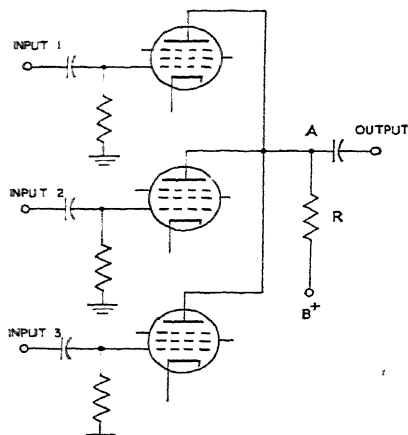


FIG. 89. Rossi coincidence circuit. A threefold coincidence arrangement is shown but may be extended to an  $N$ -fold circuit by placing  $N$  identical input stages in parallel with the tube plates connected to a common resistance  $R$  as shown. All tubes are normally conducting. Any fraction of the coincidence set may be made nonconducting by negative input pulses without altering appreciably the voltage at  $A$  provided at least one tube remains conducting. If, however, all  $N$  tubes are cut off simultaneously, their combined resistance becomes high and a large pulse is produced at the output  $A$ . [*B. Rossi, Nature, 125, 636 (1930).*]

actuate a mechanical register. For most registers available for counter circuits the output pulse should be 200 to 300 volts at 16 to 50 ma. The ideal output pulse shape is rectangular with a width of the order of 0.01 sec. This can usually be achieved by preforming the pulse before it enters the final power output stage.

Registers commonly used in counter circuits consist of a light moving armature actuated by the magnetic field of an inductance placed in the plate circuit of the output stage. Through a ratchet mechanism connected to the armature, each pulse is indicated by unit movement on some kind of dial used as an indicating device.

The maximum counting rate of a register depends on the inertia of the mechanism and the driving power. Although some registers will indicate as

many as 100 or more uniformly spaced pulses per second, considerable error is introduced in attempting to register random counts at this rate. If the pulse width or the register resolving time is of the order of 0.01 sec, two or more pulses occurring within an interval equal to or less than this are counted

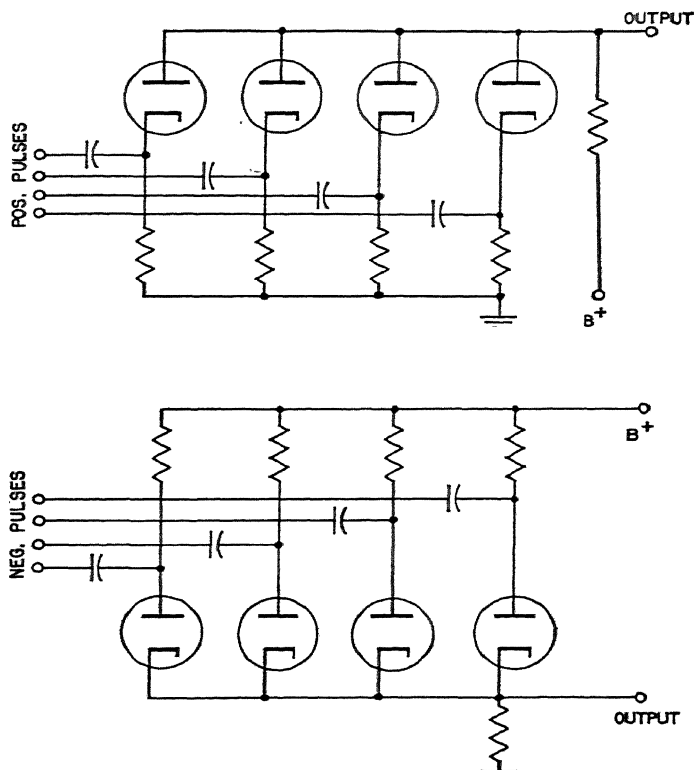


FIG. 90. Diode coincidence circuit. [B. Howland, C. A. Schroeder, and V. P. Shipman, *Rev. Sci. Instruments*, 18, 551 (1947).]

as one and therefore result in considerable loss of counts at high rates.

Typical recording circuits are shown in Figs. 86, 87, and 88.

**10.25. Coincidence Circuits.** Coincidence circuits are electronic discriminating devices that permit a count to be registered only when a predetermined number of counter tubes in the circuit discharge simultaneously, or more exactly, within a short time interval, usually of the order of the

counter-tube resolving time. Typical coincidence circuits are shown in Figs. 89 and 90.

If the polarity of the pulses from one or more counter tubes in a circuit is reversed before being mixed with simultaneous pulses from the remainder

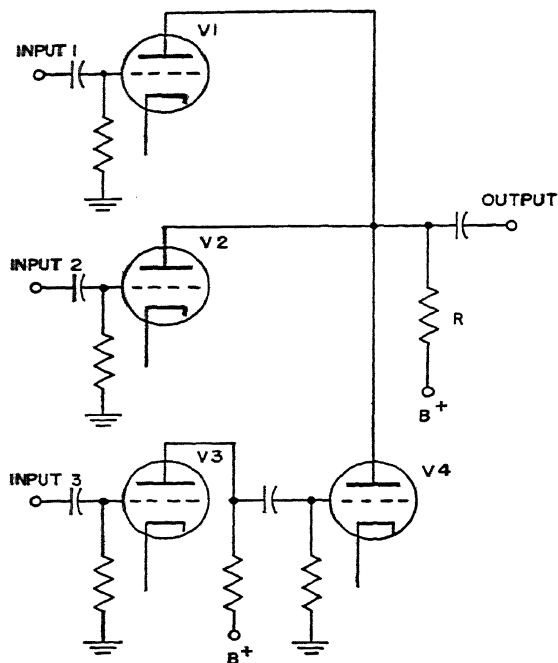


FIG. 91. Anticoincidence circuit. A pulse at input 3 is reversed in polarity in  $V_4$  and will cancel pulses at inputs 1 and 2 if they occur simultaneously with a pulse at input 3.  $V_1$ ,  $V_2$ , and  $V_3$  are normally conducting whereas  $V_4$  is normally nonconducting. The resistance  $R$  is chosen to give the correct total plate current for  $V_1$  and  $V_2$ . [G. Herzog, *Rev. Sci. Instruments*, 11, 85 (1940).]

of the tubes, such counter tubes form an anticoincidence set. Simultaneous discharges in the coincidence and anticoincidence sets cancel in the mixing stage and are therefore not registered as a count. Furthermore, independent discharges of the anticoincidence set are clipped at some stage in the counter circuit, thus permitting only independent discharges of the coincidence set to register as counts.

Examples of such circuits are shown in Figs. 89, 90, and 91.

**10.26. Counting-rate Meter.** The counting-rate meter is a circuit developed for the purpose of providing an output, usually a d-c voltage, which is

proportional to the average rate of incidence of random or periodic pulses [25-28]. A successful counting-rate meter circuit is shown in Fig. 94 and described in principle below [27,28].

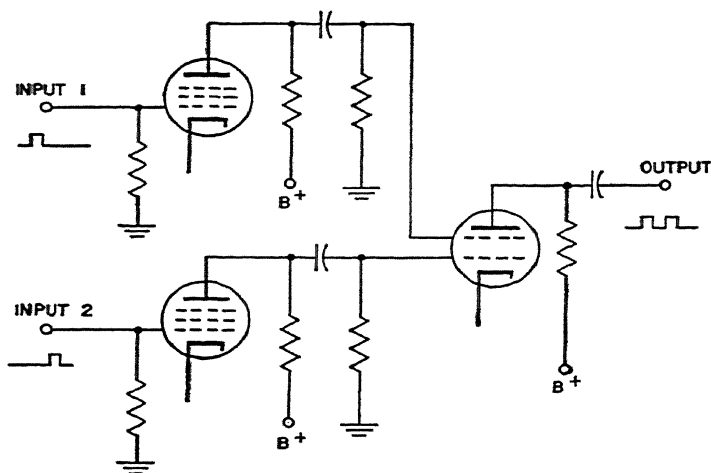


FIG. 92. Pulse-mixing circuit.

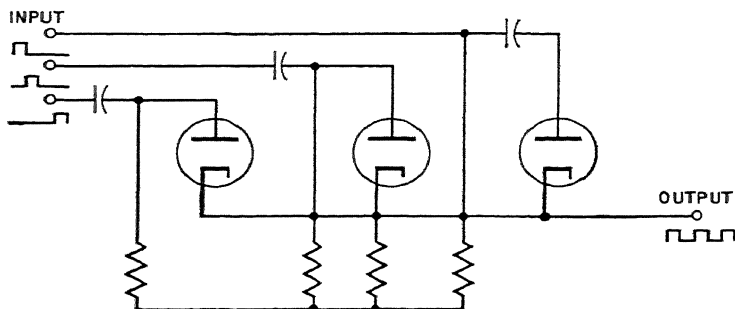


FIG. 93. Diode pulse-mixing circuit. [B. Howland, C. A. Schroeder, and V. P. Shipman, *Rev. Sci. Instruments*, **18**, 551 (1947).]

Amplified pulses from a counter are fed into a conventional multivibrator which serves as a pulse equalizer by providing an output pulse uniform in size and shape and positive in polarity for each input pulse. The equalized pulses are then impressed on the grid of the vacuum tube of the integrating circuit which contains a resistive-capacitive tank circuit in series with its plate. The d-c voltage developed across the RC circuit is proportional to

the average pulse rate and, hence, a vacuum-tube voltmeter included in the circuit may be calibrated directly in terms of average counting rate. Alternatively, automatic recording devices can be inserted when continuous records are desirable.

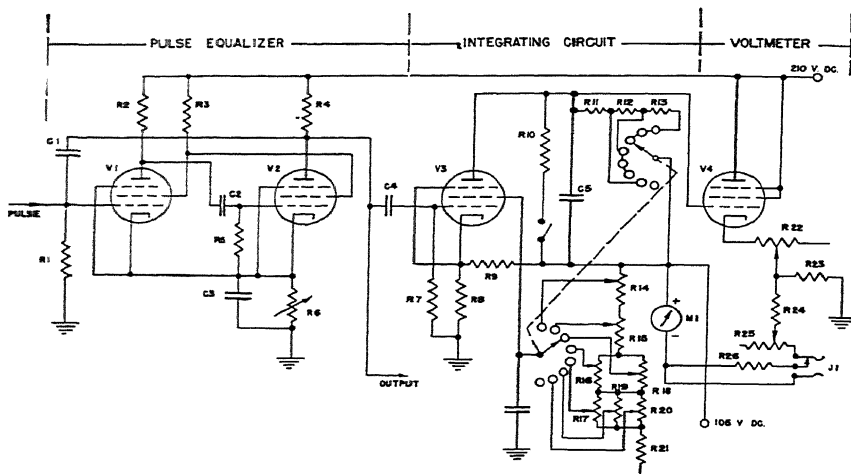


FIG. 94. Typical counting-rate meter. [A. Kip, *et al.*, *Rev. Sci. Instruments* **117**, 323 (1947).]

$R_1, R_5, R_{10} = 100,000$  ohms

$R_2, R_4 = 250,000$  ohms

$R_3, R_{22} = 300,000$  ohms

$R_6, R_{25} = 2,000$  ohms

$R_7 = 500,000$  ohms

$R_8 = 3,300$  ohms

$R_9 = 30,000$  ohms

$R_{11}, R_{12}, R_{13} = 10$  megohms

$R_{14} = 50,000$  ohms

$R_{15} = 25,000$  ohms

$R_{16}, R_{17}, R_{18}, R_{19}, R_{20} = 15,000$  ohms

$R_{21} = 8,500$  ohms

$R_{23} = 12,000$  ohms

$R_{24}, R_{26} = 1,000$  ohms

$C_1, C_2 = 100$   $\mu$ fd

$C_3 = 0.5$   $\mu$ fd

$C_4 = 200$   $\mu$ fd

$C_5 = 2$   $\mu$ fd, polystyrene

$V_1, V_2, V_3 = 6SJ7$

$V_4 = 6AC7 - GT/G$

$M_1 = D-C$  Voltmeter

$J_1 =$  Phone jack for recording milliammeter

In principle the integrating circuit establishes an equilibrium voltage across the RC tank circuit which is directly proportional to the counting rate. Equilibrium is reached when the charge leakage rate through the high resistance of the tank circuit is equal to the rate of charge added by the pulses. In general, equilibrium is established after several decay half-times of the RC circuit. Starting from zero time, equilibrium is reached [27] when

$$t_0 = RC \left( \frac{1}{2} \log 2nRC + 0.394 \right) \quad \text{sec}$$

where  $n$  = number of pulses per unit time

The statistics of this type of counting-rate meter depend on the fact that the reading at any instant is influenced by all previous pulses weighted exponentially according to their elapsed time. The absolute probable error of a single reading is given by [27]

$$= 0.477 \left( \frac{n}{RC} \right)^{1/2}$$

If a continuous recording is made, the probable error may be considerably reduced. From a record taken over a time  $T$  the mean value of  $n$  is determined by drawing a line through the trace which divides the fluctuations into equal areas on both sides. The probable error of the mean is [27] then

$$r(T) = r \frac{(1 + 2T/RC)^{1/2}}{(1 + T/RC)}$$

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## CHAPTER 11

### PROPORTIONAL COUNTERS

**11.1. General Features and Use.** The proportional counter is a variable gas-amplifying diode, in general, similar to a Geiger-Müller discharge tube in construction and materials. The particular usefulness of this form of counter derives from the nearly linear relation between the output pulse size from the counter and the total ionization produced initially in the counter by a charged particle. With appropriate electronic circuits, it is possible to discriminate between kinds of particles traversing the counter and frequently between similar heavy particles of different energies. Thus, in decreasing order of magnitude, definite pulse size for a particular counter can be associated with fission fragments, alpha particles, protons, and electrons. The property of discrimination can be used to advantage for reducing the observed background from light particles and gamma radiation when low intensities of heavily ionizing particles are measured, by adjusting the counter circuit to amplify and register only pulses larger than a predetermined size.

The design of proportional counters depends mainly upon the radiation to be measured. For most applications three forms are commonly used:

1. "Bell" shaped counters with tungsten wire anodes and with thin windows for detecting alpha and beta radiations.
2. Cylindrical glass or metal tubes with coaxial tungsten wire anodes for neutrons and cosmic radiations.
3. Point counters [1] similar to "bell" counters but with a spherical metal anode 1 to 2 mm in diameter supported near the counter window. The sensitive region of these counters lies in only a small volume surrounding the ball anode.

In addition a design developed by Zipprich [2], consisting of two parallel plate electrodes and an accelerating grid placed between the plates, is less often used but possesses the advantage of a well-defined sensitive region.

Filling gases for proportional counters need not contain polyatomic, or quenching, gases although the inclusion of such gases provides greater stability, particularly for large pulses. The gases most frequently used are a mixture of argon, 10 to 25 per cent, and methane, 75 to 90 per cent, or various mixtures of nitrogen, methane, ethane, and ether. Pressures commonly used vary from a few centimeters to half an atmosphere.

The operating voltage for a proportional counter is determined largely by the size pulse required for the particular radiation to be measured. Before the counter can be used, however, it is necessary to determine the proportional region by plotting the counting rate against anode voltage. As shown in Fig. 77, the region for proportional pulse size lies between the starting voltage at which the first counts are detected and the beginning of the transition to the Geiger-Müller region.

**11.2. Theory of Operation.** When certain simplifying assumptions are made which appear to be valid for most proportional counters, the formation of a voltage pulse proportional to the initial number of ion pairs is adequately explained in terms of a Townsend avalanche [3,4,5]. It is assumed that (1) the probability of electron photoemission is negligibly small; (2) the probability for secondary electron emission by positive ions collected at the cathode is negligibly small; (3) negative ions such as  $O^-$ ,  $F^-$ , and  $Cl^-$  are not sufficiently numerous to produce an appreciable lag in the counter action; (4) recombination of positive ions and electrons within the gas is negligible. From the  $N$  initial ion pairs produced by an incident charged particle, the electrons are collected at the anode in a time of the order of a few microseconds while the positive ions formed remain essentially stationary during this time. Within a distance of several mean free paths from the anode, electrons acquire sufficient energy between collisions in the strong electric field to produce additional ion pairs and hence a Townsend avalanche. The avalanche at this point differs from the Geiger-Müller counter discharge in that the descendants of a single electron multiply to a smaller and, on the average, constant number which is a function of the operating voltage. The factor by which an electron multiplies itself in the avalanche, called the amplification  $A$ , may have a value up to a maximum of  $10^4$ , although generally it is of the order of  $10^3$  or less. Within the range from 1 to  $10^4$ , its value can be chosen arbitrarily by the proper choice of operating voltage, a factor that permits some control over the pulse size which may be desired. When the amplification factor increases beyond  $10^3$  or  $10^4$  owing to a high operating voltage, the pulse size is no longer strictly proportional and some of the characteristics associated with the Geiger-Müller discharge make their appearance. Following the completion of the Townsend avalanche the residual positive ion cloud drifts slowly to the cathode. However, during the dead time of roughly  $10^{-4}$  sec, it still exists as a space charge sufficiently near the anode to reduce the effective electric field and thus prevent a second avalanche which might be initiated by a charged particle arriving within this time.

The total charge collected by the anode is approximately  $Q = eAN$ , where  $e$  is the electronic charge. With a total capacitance of  $C$  for the

counter and its immediately connected circuit, the peak pulse voltage is then

$$V = \frac{eAN}{C}$$

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## CHAPTER 12

### IONIZATION CHAMBERS

**12.1. Description.** The ionization chamber may be described in the most general terms as a gas-filled chamber in which a constant electric field is maintained by a set of electrodes for the collection of ions formed by incident ionizing radiation. In principle, the charge collected should exactly equal the total ionization formed in the sensitive volume of the chamber by the incident radiation. This linear relationship between ionization and collected charge is the most important distinguishing characteristic of the ionization chamber. It is necessary, therefore, that the field strength be high enough to collect the ions formed before appreciable recombination and diffusion of ions has taken place in the gas, yet not so high as to cause the formation of additional ions by multiplicative processes as the charge drifts to the collecting electrodes (see Fig. 78). The useful operating range of potential difference (arbitrary with respect to ground potential) maintained between the electrodes can be found by observing the ionization in the chamber as a function of voltage for a constant source of radiation. Starting with zero potential difference, the detected ionization increases with voltage until saturation is attained. The ionization then changes relatively little with further increase in voltage until the field strength becomes sufficiently high to induce ion multiplication by electrons as they drift to the anode. The operating voltage should, of course, lie in the plateau region. In practice, the field strengths used vary from 20 to 500 volts per cm and usually are not critical.

Two methods for the detection of the ionization in the chamber may be used: either measurement of the constant voltage produced by the flow of collected charge through a resistance, or measurement of the rate of change of voltage as charge is collected. With a high resistance  $R$  connected between the electrodes, the change in potential difference between them after a time  $t$ , assuming the rate of ionization is constant, is

$$\Delta V = V_o - V = neR(1 - e^{-t/RC}) \quad \text{volts}$$

where  $V_o$  = operating voltage

$n$  = number of ions formed per second

$e$  = electronic charge

$C$  = capacitance of electrode system

When  $t \gg RC$ , the voltage change reaches a constant value  $\Delta V = neR$ . The capacitance  $C$  can usually be made very small,  $\sim 10$  to  $100 \mu\text{mfd}$ , so that the

time required to reach the steady state is conveniently short,  $< 1$  sec, even for the high resistances necessary to make  $\Delta V$  easily measurable. For ionization chambers in which pulses are detected, it is usually necessary to make  $RC$  less than 0.001 sec. On the other hand, if  $R$  is removed or, more

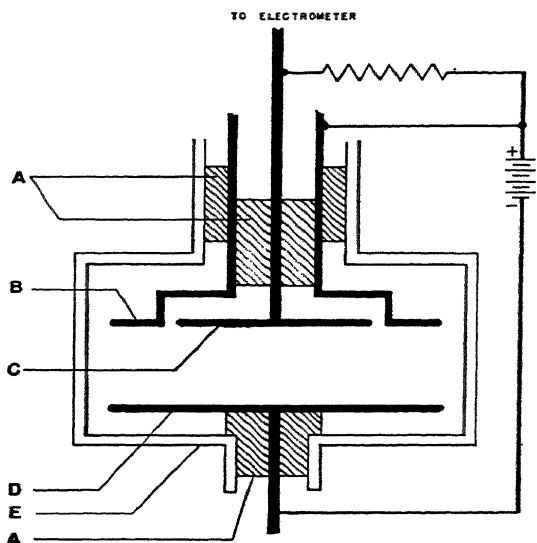


FIG. 95. Schematic diagram of the parallel-plate ionization chamber. The essential components of these chambers are *A*, low-leakage insulators between the electrodes; *B*, guard ring, maintained at the same potential as the collecting electrode; *C*, collecting electrode (anode); *D*, high-voltage electrode for maintaining the charge collecting field (cathode); *E*, chamber which may be designed to withstand high or low pressures or may serve merely to shield against external fields.

exactly, made very large,  $\sim 10^{13}$  ohms, it is convenient to measure the time rate of change of  $\Delta V$  since

$$\frac{d(\Delta V)}{dt} = \frac{ne}{C} e^{-t/RC} \quad \text{volts/sec}$$

or in an interval of time  $t \ll RC$

$$\Delta V = \frac{ne}{C} t \quad \text{volts/sec}$$

and the linear increase in  $\Delta V$  with time is measured. This method is usually used only when the smallest ion currents,  $ne < 10^{-14}$  amp, are to be detected.

The details of the construction of an ionization chamber depend to some extent on the type of radiation to be detected, but more often its design and

construction materials are determined by the particular circumstances of the measurements. Despite great variations in design, the chambers are, in most instances, built around two basic forms of collecting electrode geometry. The first is a parallel-plate condenser shown in Fig. 95. The central portion of one plate of the condenser is highly insulated and connected directly to the detecting instrument as well as to some means for charging it to the desired potential, often ground potential, either through a charging (or grounding) switch or a high grid resistance if a vacuum-tube amplifier is used. The remainder of the plate bounding the collecting electrode serves as a guard ring and is maintained at the same potential to define accurately the sensitive volume of the chamber and to prevent leakage of current across

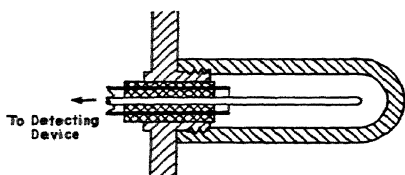


FIG. 96. Thimble ionization chamber.

the insulator supporting the collecting electrode. The opposite plate of the condenser is maintained at the potential necessary to provide the requisite field strength between the electrodes. The second form of chamber frequently used consists of a cylindrical anode (or cathode) within which a highly insulated rod-shaped collecting electrode is coaxially mounted, as shown in Fig. 96.

The functions of the guard ring, which is maintained at the same potential as the collecting electrode, are especially important in chambers intended for measurement of weak ionization. The least important of these is to define the sensitive volume. Its more important functions are (1) prevention of leakage currents across the insulator supporting the collecting electrode by maintaining zero potential gradient, (2) prevention of insulator stresses due to potential gradients, since polarization and relaxation of the insulator may induce voltages comparable to those produced by the ions, and (3) elimination of steep potential gradients near any part of the collecting electrode system.

All ionizing radiations can be detected with appropriate forms of chambers. Heavily ionizing particles such as protons, alpha particles, and fission fragments produce ionization of sufficient intensity to be detected and counted as separate events using the same techniques and external counting circuits as for Geiger-Müller and proportional counters. Less strongly ionizing radiations such as gamma rays and beta particles cannot be detected and counted as separate events but must be measured by the average charge collected per unit time at the electrodes.

Chambers designed for radiation detection by the time rate of accumulation of charge normally will operate satisfactorily when filled with any gas of low molecular weight, including those gases, such as oxygen, water, carbon

monoxide, carbon dioxide, and the halogens, which form negative ions. Air at atmospheric pressure is the most frequently used gas. Chambers operated at high pressures for the purpose of contracting the range of the radiation are often filled with argon or krypton since these gases have greater stopping power and large cross sections for ionization. Pulse chambers, on the other hand, frequently exhibit erratic performance and provide poor pulse shapes when filled with negative ion-forming gases. Usually they are operated with nitrogen, argon, or methane at atmospheric pressure, and in many instances it is convenient to allow the gas to stream through the chamber. This latter method is especially useful when the source of radiation is placed in the chamber and is frequently changed as in measuring radioactive samples.

**12.2. Applications.** The ionization chamber has found more extensive use and more diverse adaptations than any other device for measuring ionizing radiations. Its simplicity and comparative ruggedness are factors to be considered to its advantage but, most important, the ionization chamber lends itself to measurements of radiation under conditions that cannot be duplicated with other means for detecting ionization. In general, applications of the chamber may be divided into two categories. Although these categories are somewhat superficial, there exists an important physical distinction between them. The first includes measurements of radiations that have ranges comparable to or smaller than the dimensions of the gas volume, and the second applies to penetrating radiations that have ranges very much greater than the dimensions of the chamber.

The first category involves measurements on short-range charged particles and slow neutrons. The ionization produced in the chamber can usually be interpreted unequivocally in terms of the number of primary particles or as relative activity of the source of radiation. The design of chambers for this purpose is largely a matter of expediency, consistent with special features of the application and acceptable instrument designing. The source of primary radiation, because of the comparatively short range of charged particles, must be placed within the chamber to permit the greatest possible portion of the range to lie in the sensitive gas volume. In the case of slow neutrons the source may be boron or a fissionable material (see Sec. 10.10). In nearly all cases where the source material is solid, it can be deposited in a thin film on one of the electrodes or on a suitable support that is mounted adjacent to the sensitive volume of the chamber. For the highest efficiency the film thickness should equal approximately the range of the particle in the substance. A somewhat greater ionization efficiency and sensitivity can be achieved when the substance can be introduced as part of the chamber gas since self-absorption is then eliminated and the geometrical efficiency is increased to very nearly 100 per cent. Thus boron trifluoride is most commonly used for detecting slow neutrons, and carbon dioxide and hydrogen-

filled chambers are among the most sensitive instruments for measuring  $C^{14}$  and  $H^3$ , respectively.

The second category of applications involves gamma radiation and fast neutrons but may also include charged particles with very high energies such as those encountered in cosmic rays and from high-energy accelerators. The interpretation of the measured ionization in this case is not always obvious since the chamber walls perform an important function in affecting the observed intensity of ionization.

When the range, or more correctly the half-value thickness, of the primary radiation in the gas contained in the chamber is very much greater than the chamber dimensions, the ionization produced and measured is, in the most general case, the sum of the ionization produced in the gas volume by the primary radiation plus that produced by recoil and scattered particles from the walls. With chambers of wholly arbitrary dimensions and materials it is difficult to assign values for the energy and flux of primary radiation from the measured values of the ionization because it is nearly impossible to ascertain what fraction of the ionization is contributed by secondary corpuscular radiation produced in the walls. Only when the chamber has been calibrated for a specific type of primary radiation and for a particular energy can the indicated ionization be interpreted in terms of dose or as energy flux.

The contribution of the walls to the observed ionization depends principally on the following factors:

1. Atomic number of wall materials.
2. Thickness of the wall relative to the range of secondary corpuscular radiation.
3. Dimensions of the chamber relative to range of secondary corpuscular radiation.
4. Attenuation of primary radiation effected in the walls.
5. In the case of neutrons, possible nuclear reactions.

The only simple conclusion that can be drawn concerning these factors is that the maximum wall effect is attained when the wall thickness equals the range of secondary particles produced in it and the material is chosen to give the highest yield of particles; for gamma rays these are materials possessing the highest electron densities such as copper and lead, and for fast neutrons these are hydrogenous substances.

In two special cases the foregoing difficulties in evaluating the physical significance of the measured ionization reduces to results that permit simple interpretation. In the first case, consider a collimated beam of primary radiation passing between but not striking the electrodes of a parallel plate chamber. If the walls are placed at large distances compared to the range of the secondary particles in the chamber gas, then the ionization produced in the sensitive volume corresponds exactly to the energy absorbed in the



gas. If the gas is air, the average energy absorbed per ion pair is known for various radiations and the total energy absorbed in the sensitive gas volume is readily calculated.

The second special type of ionization chamber provides a means for measuring the amount of energy absorbed from the primary radiation in traversing solid substances and therefore is of the most profound importance to dosimetry and to the absolute measurement of energy flux. For this reason it is described here in somewhat greater detail.

It has been shown by Gray [8,9,13] that for a small air-cavity surrounded by a medium  $M$  the energy absorbed in  $M$  per unit volume per second from the primary radiation is related to the ionization observed in the cavity by the expression

$$E = SJW \quad \text{ergs or mev/cc/sec}$$

where  $E$  = energy absorbed per unit volume per second in medium  $M$  (wall material)

$S$  = ratio of stopping power for secondary particles in medium  $M$  to stopping power of the gas

$W$  = average energy absorbed from secondary particles to form one ion pair in the gas (see Sec. 3.9)

$J$  = number of ion pairs formed in the cavity per unit volume per second

The quantity  $S$  is the ratio of energy loss per unit length of path in the medium  $M$  to that in the gas. This may be expressed most conveniently in terms of the atomic stopping powers  $B$  and densities  $\rho$  of the two media. Thus,  $S = \rho_m B_m / \rho_g B_g$ . In many instances the values of  $B$  are not known with certainty, but if the atomic composition of the gas can be made similar to that of  $M$ , then  $B_m = B_g$  and  $S$  is given directly as the ratio of only the densities.

The simple but important relation above is valid only when the cavity ionization chamber meets the following physical conditions:

1. The dimensions of the cavity must be smaller than the average range of the secondary corpuscular radiation in the gas contained in the cavity.
2. The wall thickness of material  $M$  surrounding the cavity must be equal to or greater than the maximum range of the secondary particles in  $M$ .
3. The primary radiation should not be appreciably attenuated in traversing the chamber.
4. The relative stopping power must be independent of the velocity of the secondary particles.

It is apparent from these conditions that for the equation to be valid the intensity of secondary corpuscular radiation passing through the cavity must exactly equal its intensity within the wall material when it is in radiative equilibrium with the penetrating primary component.

The significant properties of chambers for which the formula and conditions above are valid are [9] (1) for sufficiently small chambers the ionization per unit volume is independent of the size of the cavity; (2) the ionization is directly proportional to the gas pressure; (3) for the same primary radiation the ionization in chambers with different wall materials is proportional to the energy absorbed per unit volume of wall material and inversely proportional to its stopping power.

It is apparent that size is a distinguishing and crucial feature of the cavity chamber, but it is not always clear just how small the dimensions must be since the minimum range of recoil electrons in the case of gamma rays, and recoil protons from neutrons, is essentially zero. In principle the size should be "infinitesimal," but in practice cavity dimensions of the order of millimeters are usually permissible without introducing appreciable error. In any actual application this can be tested with cavities of progressively larger dimensions. The largest cavity that still indicates a constant ionization per unit volume can be regarded as sufficiently small for that particular wall material and radiation.

The exactness of the cavity formula above is also influenced by two factors in addition to finite cavity size. These are (1) the ionization produced in the gas by the primary radiation directly is, in general, not equal to that which would be produced in an equivalent volume of wall material, and (2) the presence of the cavity alters slightly the energy and directional distribution of the secondary corpuscular radiation. These factors are only significant when the constituents of the wall material and gas differ markedly in atomic number, such as in the combination of air and lead. In practice their influence can be ascertained by observing the ionization as a function of pressure. If pressure and ionization are proportional, the cavity formula can be assumed to be valid. The most accurate results are to be expected with wall material and gas of the same atomic composition—a combination that is sometimes difficult to achieve.

In applications of the cavity chamber to problems of dosimetry the wall materials usually consists of compositions containing several kinds of atoms in proportions similar to the composition of tissue, and in addition, the primary radiation frequently is heteroenergetic. If these conditions are to be taken into account explicitly, the cavity formula, though still valid, is modified [10] into the form

$$E = \Phi N \sum_i f_i \overline{\gamma_i} \quad \text{ev/cc/sec}$$

where  $\Phi$  = flux of primary radiation, ev per cm<sup>2</sup> per sec

$N$  = number of atoms per cc of wall material

$f_i$  = fraction of atoms of the kind  $i$

$\gamma_i$  = average energy of recoil particles from atom of kind  $i$  in units of primary radiation energy

$\sigma_i$  = collision cross section for atom of kind  $i$

The bar over the product  $\gamma\sigma$  indicates its average value taken over the energy spectrum of the primary radiation. When applied to gamma rays in the energy range where only Compton scattering is important, the formula can be used in a somewhat simpler form.

$$E = N_e \bar{\Phi} \gamma_e \sigma_e \quad \text{ev/cc/sec}$$

where  $N_e$  = number of electrons per cc of wall material

$\gamma_e$  = average energy of recoil electrons

$\sigma_e$  = electronic scattering absorption coefficient (see Sec. 2.3)

A special form of cavity chamber has been extensively used by Failla [11] for the investigation of the dose delivered by x- and gamma radiation at small depths in tissuelike substances. Its importance derives from the fact that it provides a method for measuring the primary radiation energy absorbed per unit volume of solid at depths from the surface corresponding to the transition region before radiative equilibrium is reached between the primary and secondary radiation. The usual form of the chamber consists of a small shallow chamber, less than 1 mm deep, machined in a block of material of the desired atomic composition. Extremely thin coats of graphite serve as electrodes since, though sufficient to conduct charge, they do not influence the ionization. The dose curve throughout the transition region can now be plotted by successively decreasing the wall thickness through which the radiation enters normal to the chamber axis. When sufficient points are determined, the curve can be readily extrapolated back to zero wall thickness, thus giving the dosage rate or the energy absorbed at the surface and at any depth in the substance. The surface dose indicated by this method is the true dose since the contributions from scattered primary and secondary radiation appear implicitly in the measurements.

**12.3. Charge-measuring Instruments.** The charge collected at the electrodes of an ionization chamber requires for its measurement instruments of the highest possible sensitivity. In most instances the rate of charge formation is smaller than  $10^{-10}$  amp and may be as little as  $10^{-16}$  amp. It is evident, therefore, that both the chamber and the charge-measuring device must be not only sensitive but must be made relatively free from extraneous influences such as excessive insulator leakage, stray fields, thermal coefficients, contact potentials, voltages due to thermal agitation, and fields induced by insulator stresses which can produce spurious responses of the same order of magnitude as the charge to be measured. In all detecting instruments employed with ionization chambers, the physical quantity

measured is a small potential difference produced by the collected charge. Although the millivolt is the absolute quantity measured directly, such instruments may be calibrated in terms of roentgens, curies of source material, or in arbitrary units of radiation intensity.

The instruments most frequently adapted to ionization-chamber measurements are summarized below.

*a. Quartz-fiber Electroscope.* Of the numerous types of electroscopes that have been used for radiation detection, the form developed by Lauritsen is the most frequently used in this country. It consists of a metal-coated

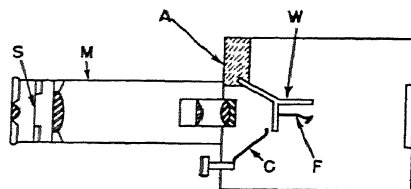


FIG. 97. Schematic diagram of Lauritsen electroscope. *A*, amber insulator for supporting fiber assembly; *C*, switch for charging fiber; *F*, metal-coated quartz fiber with "T" fiber mounted at free end; *M*, compound microscope; *S*, scale for reading displacement of fiber; *W*, stiff wire support for fiber.

quartz fiber, 6 mm long and 0.005 mm in diameter, mounted parallel to a rigid wire and connected mechanically to it, as shown in Fig. 97. An aluminum shield covers the fiber assembly to serve as the cathode and to protect the fiber from damage, air currents, and dust. The displacement of the end of the fiber is observed with a compound microscope in which an accurately ruled scale is mounted in the focal plane of the eyepiece. Although this type of electroscope is designed to be used independently, the microscope and

fiber assembly can be mounted on an ionization chamber with the fiber connected directly to the collecting electrode.

The fiber is initially displaced by charging it with a potential of approximately +100 volts before a measurement is to be made. Then, as ions of the opposite charge are collected, the fiber drifts back at a rate proportional to the rate at which ions are produced in the sensitive region surrounding the fiber or in an ionization chamber connected to it. Small displacements of the fiber are very nearly proportional to the number of ions collected, and a linear relationship can usually be assumed if the displacement is not large.

Normally, the background activity from cosmic rays and alpha particles gives a drift rate of five to ten scale divisions per hour.

*b. Moving-vane Electrometer* [1]. The most suitable electrometers of this type for ion detection are those with a metal-coated quartz fiber mounted on a torsion fiber in the gap between constant potential electrodes. Both the Hoffman [2] and Lindemann [3] forms have been widely used with ionization chambers, but at the present time the Lindemann electrometer generally is preferred (see Fig. 98). It is highly sensitive, its calibration is independent of orientation, and it can be made portable.

With the appropriate electrical circuits the rate of charge collection can be read directly in terms of scale divisions per second, or as a null instrument it can be calibrated in terms of millivolts per second required to maintain the fiber at the zero position. The latter method is preferable when the highest accuracy is desired.

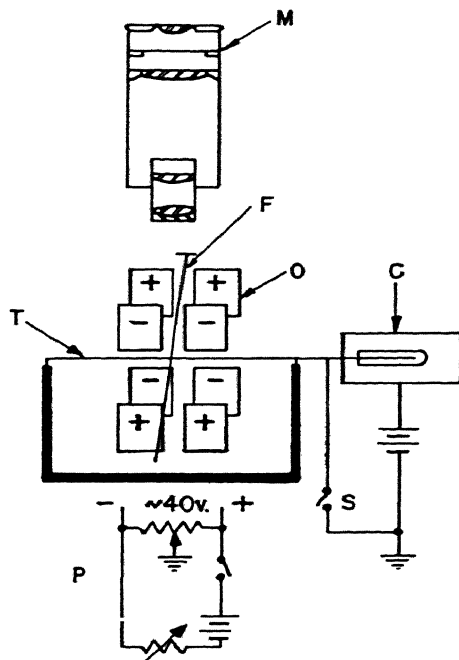


FIG. 98. Schematic diagram of Lindemann electrometer and external circuits. C, ionization chamber; F, metal-coated quartz fiber with "T" fiber at one end; M, compound microscope; O, octants of electrometer; P, circuit for applying accurately known voltage to octants; S, shorting switch to discharge collecting electrode and fiber; T, torsion fiber.

*c. Vacuum-tube Electrometer.* The voltage produced by leakage of the collected charge across a high resistance of the order of  $10^{11}$  ohms can be amplified by an electrometer-connected vacuum tube similar to the method described in Sec. 9.11. Several vacuum tubes such as the FP-54 and the Victoreen VX-41 [4,5] have been designed specifically for this purpose.

This method is applicable to currents as low as  $10^{-15}$  amp. The sensitivity can be further extended, to about  $10^{-17}$  amp, if the resistance is removed and the charged is allowed to accumulate. The voltage developed on the grid of the vacuum tube then depends upon the total capacitance of the grid

circuit and the total charge collected. It is important in this case to choose electrometer tubes with extremely low grid currents and to use insulation of the highest order.

Direct-current amplifiers have had only limited use with ionization chambers partly because of the technical difficulties in their construction and operation and partly because of their susceptibility to microphonics, long period drift and high noise level as compared with other detecting devices. However, in recent years the use of 100 per cent feedback circuits (current amplifiers) have eliminated many of their former shortcomings and are finding more extensive applications to the detection of very small currents.

*d. Vibrating-reed Electrometer.* The vibrating reed is a variable capacitor for converting the d-c potential developed across a capacitor by the collected charge from the ionization chamber into an oscillating voltage that can be

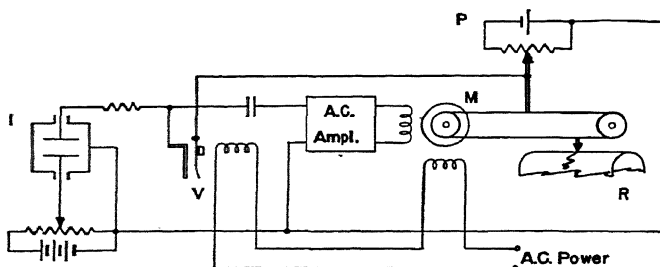


FIG. 99. Schematic diagram of vibrating-reed electrometer. *I*, ionization chamber, *M*, two-phase motor; *P*, slide-wire potentiometer operated by motor *M*; *R*, chart paper and recording pen; *V*, vibrating-reed capacitor and driving coil.

amplified by conventional a-c amplifiers [6,12]. With a constant charge on the collecting system, the voltage across the capacitor is

$$V = \frac{Q}{C(t)} = \frac{Q(d + a + a \sin \omega t)}{kA + C_0(d + a + a \sin \omega t)}$$

where  $Q$  = charge on electrode system

$C_0$  = capacitance of collecting electrodes

$k$  = dielectric constant

$A$  = area of reed

$d$  = minimum separation of vibrating reed electrodes

$a$  = amplitude of vibration of reed electrodes

$\omega$  = vibrational frequency of reed

The oscillating component of the reed potential, which increases as charge accumulates, is applied to the input of a conventional linear a-c amplifier. After amplification it is rectified and the resulting d-c voltage measured by a recording potentiometer which automatically and continuously applies to

the vibrating reed a reverse voltage just sufficient to balance the signal voltage. This feature provides a considerable advantage in taking measurements since the slope of the recorded trace is proportional to the ionization rate. Further, the incidence of an alpha particle in the chamber produces an abrupt deflection in the slope of the line, thus allowing the background of alpha particles to be fully accounted for separately in each measurement without resorting to statistical methods.

The vibrating-reed electrometer is less susceptible to microphonics and to those factors which in other forms of electrometers frequently cause long period drift and high noise level [6]. It is one of the most useful instruments for detecting small currents because it combines very high sensitivity with great stability and can be used under the most adverse physical conditions.

*c. Alternating-current Amplifiers.* Heavily ionizing particles such as protons, alpha particles, and fission fragments produce sufficient ionization to be detected in ionization chambers with a-c pulse amplifiers and counted by conventional recording circuits. The voltage developed across a resistance of  $10^8$  to  $10^{10}$  ohms is impressed on the grid of a vacuum tube operated usually as a cathode follower, *i.e.*, with a cathode resistor of 10,000 to 50,000 ohms and the output signal taken from the cathode. This is followed by one or two stages of amplification and possibly a cathode follower output stage leading to the counting circuits. Considerable precautions must be taken with preamplifiers of this sort to ensure good insulation of the grid circuit of the first tube and generally to provide insulation and shielding against vibration and external electromagnetic fields. Miniature tubes of the types 6AK5, 9001, 9002, 945, etc., have proved to be highly satisfactory for this application [7].

**12.4. Statistics of Measurements.** The probable error of measurements with an ionization chamber recording discrete events is estimated by the same procedure used for other counting methods and is given in detail in the section on statistics for Geiger-Müller counters. Chambers for detecting beta, gamma, and cosmic radiation, on the other hand, require the measurement of a continuous variable which may be either scale division per second or millivolts per second. The procedure is precisely the same, however, when the terms are redefined.

Assuming only cosmic rays and alpha particles from the chamber walls to be responsible for the observed background, the standard deviation of a single measurement of the background charge taken over a time  $t_b$  is

$$\sigma_b = \frac{1}{\sqrt{t_b}} (a^2 A + b^2 B)^{1/2} \quad \text{ion pairs/sec}$$

where  $a$  = average number of ion pairs produced in the chamber per alpha particle

$b$  = average number of ion pairs produced in the chamber per cosmic ray

$A$  = average number of alpha particles per unit time

$B$  = average number of cosmic rays traversing chamber per unit time

In the presence of the radiation being measured, the standard deviation for a single measurement of the radiation plus background over a time  $t_r$  in terms of charge units is

$$\sigma_r = \frac{1}{\sqrt{t_r}} (r^2 R + a^2 A + b^2 B)^{1/2} \quad \text{ion pairs/sec}$$

where  $r$  = average number of ion pairs produced in the chamber per incident particle or ray

$R$  = average number of events per unit time in chamber

The total standard deviation, therefore, is

$$\sigma = (\sigma_b^2 + \sigma_r^2)^{1/2} \quad \text{ion pairs/sec}$$

Alternatively, a measurement can be expressed in millivolts per unit time by

$$v_b = \frac{10^3}{Cq} (aA + bB) \quad \text{mv/sec}$$

$$v_r = \frac{10^3}{Cq} (rR + aA + bB) \quad \text{mv/sec}$$

where  $C$  = total capacity of collecting electrode plus its external connections; *e.g.*, electrometer fiber and connections or vacuum tube grid and lead

$q$  = number of ion pairs per coulomb

The standard deviations in terms of millivolts per second are then

$$\sigma'_a = \frac{10^3}{Cq} \sigma_b \quad \text{mv/sec}$$

$$\sigma'_r = \frac{10^3}{Cq} \sigma_r \quad \text{mv/sec}$$

$$\sigma'_t = [(\sigma'_r)^2 + (\sigma'_a)^2]^{1/2} \quad \text{mv/sec}$$

When a set of  $N$  measurements have been taken, the standard deviation from their mean value  $\bar{v}$  is given by

$$\sigma = \sqrt{\frac{1}{N-1} \sum_{i=1}^N (\bar{v} - v_i)^2} \quad \text{mv/sec}$$

where  $v_i$  = values of individual determinations in units of mv per sec  
This is calculated for  $N_B$  determinations of the background and  $N_R$  deter-



minations of the measured radiation plus background to obtain the total standard deviation from the mean  $\sigma_T$ , where, as before,

$$\sigma_T = (\sigma_B + \sigma_R)^{1/2} \quad \text{mv./sec}$$

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## CHAPTER 13

### STANDARDIZATION OF RADIOACTIVE SAMPLES

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**13.1. Introduction.** Precise methods for determining the absolute intensity of radioactive samples are desirable for the uniformity and standardization of measurements and are highly important in dose determinations. In this chapter the most commonly used methods of measurement of alpha, beta, and gamma radiations are described.

The methods and units of absolute measurement have not yet been developed to an entirely satisfactory stage, and the agreement between measurements made in different laboratories is not quite perfect. Efforts toward unification, however, have been undertaken by the National Research Council which recently established a committee on radioactivity to recommend standard procedures and units in the United States.

In order to have a clear conception of the units used in radioactivity, the terminology used in this field should be mentioned. Since, except for a few units, there is no international agreement on the terminology and the magnitude of the units used in the present chapter, they will be defined according to present usage (see also Chap. 7).

The rate of disintegration of a radioactive substance is the rate of change in the number of atoms of the parent radioactive isotope per unit time. The decay usually occurs by the spontaneous emission of a charged particle and one or more quanta; the emission of two or more such radiations is, for practical purposes, considered to be simultaneous. The rate of emission of radiations from a quantity of radioactive substance is the total number of quanta and particles emitted per unit time. For example, each time a radioactive  $\text{Na}^{24}$  atom disintegrates into  $\text{Mg}^{24}$ , one beta particle and two gamma-ray quanta are emitted. The rate of emission is therefore three times the rate of disintegration of  $\text{Na}^{24}$  atoms. The unit of radioactivity is usually expressed in terms of disintegration rates, whereas dose calculations often require the knowledge of emission rates.

Conventionally the unit of activity or quantity of radioactive material is the curie. One curie is the quantity of a radioisotope which decays at the rate of  $3.7 \times 10^{10}$  disintegrations per second. (One millicurie corresponds to  $3.7 \times 10^7$  disintegrations per second; one microcurie, to  $3.7 \times 10^4$  disintegrations per second.) The curie was originally used as a measure of

the quantity of radon gas in equilibrium with one gram of radium. Later this definition of the curie was extended to include other naturally radioactive decay products of radium [1]. Finally, by general use, the unit was extended to all radioactive isotopes. Recent determinations do not give the rate of disintegration for one gram of radium as  $3.7 \times 10^{10}$  disintegrations per second. The best value is somewhat lower:  $(3.608 \pm 0.028) \times 10^{10}$  per sec. This was obtained by Kohman *et al.* [2] and corresponds to 0.652 cc of radon gas at 0°C and 760 mm Hg. There is thus a small discrepancy between the number of disintegrations adopted by international agreement for one curie and the actual number. To avoid ambiguities that may arise if the curie is used in these two different connections, Curtis and Condon recently proposed a new unit for radioactivity which they called the rutherford [3]. As defined by them, one rutherford is the quantity of radioisotope decaying at the rate of  $10^6$  disintegrations per second. This unit has certain merits inasmuch as it allows the use of the term curie for the purpose originally designated, namely, to denote quantities of radium decay products. So far, however, the unit has not found general acceptance.

**13.2. General Methods.** In most measurements one is interested in the specific activity or the quantity of radioactive isotope per gram of sample substance expressed in curies per gram. If the entire quantity, *i.e.*, every atom, of a particular element in the sample is radioactive, it is called a carrier-free (C.F.) sample. The rate of disintegration  $R$  is related to the number of parent atoms  $N$  by

$$R = \lambda N$$

where  $\lambda$  is the decay constant. If  $R$  is the unknown, it is necessary to evaluate both  $\lambda$  and  $N$ , or if  $N$  is the unknown, one must measure  $\lambda$  and  $R$ . Most of the methods of primary standardization depend on measuring these quantities. The number  $N$  can be determined either gravimetrically, as in the preparation of some primary standards of uranium, or by mass spectrographic deposition of a known amount of carrier-free isotope.  $R$  is obtained if the absolute number of particles and radiations emitted can be measured or if the quantity of the accumulated decay products is measured over a certain time interval.

**13.3. Primary Alpha-particle Standards.** Because of the accuracy with which the disintegration constants are known, the most satisfactory alpha standards are prepared from several of the long-life natural alpha-particle emitters. Uranium and  $\text{Ra}^{226}$  have been used more than others. With weighed amounts of these substances it is possible, with appropriate corrections, to count all alpha particles emitted from them in alpha-particle ionization chambers. Since the vast majority of alpha particles follow straight trajectories, have a well-defined range, and produce relatively

large specific ionization, under certain conditions each particle crossing the sensitive volume of the chamber will be recorded. Corrections for absorption and geometry can then be readily made to determine the absolute number of particles emitted. By this technique, Kovarik and Adams [4] showed that 1 gm of uranium emits a total of  $2.501 \times 10^4$  alpha particles per second.

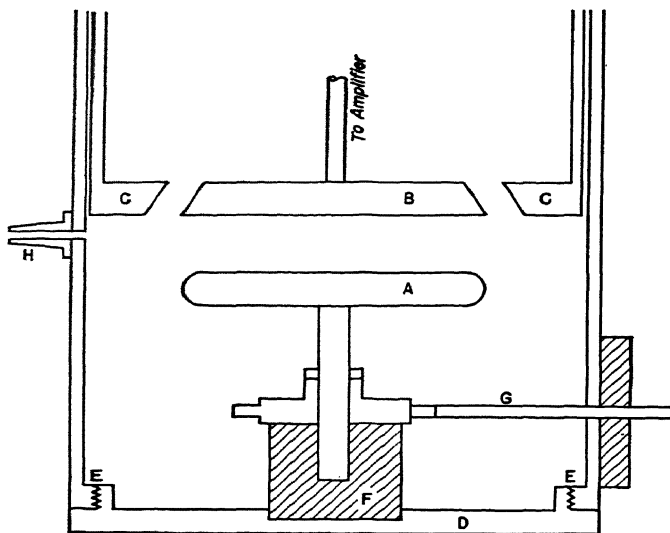


FIG. 100. Alpha-particle ionization chamber. Principal components are: *A*, high-voltage electrode on which sample is mounted; *B*, collecting electrode; *C*, guard ring; *D*, bottom plate; *E*, breach thread; *F*, amphenol insulator; *G*, high-voltage wiping contact; *H*, inlet for nitrogen or argon. [Redrawn from A. H. Jaffey, T. P. Kohman, and J. A. Crawford, U.S. Atomic Energy Commission Report MDCC-388, 1946.]

Knowing the isotopic constitution of uranium, mainly from the work of A. O. Nier [5], the disintegration rate and half-life of each of its three isotopes can be determined, as shown in Table 35.

TABLE 35. DISINTEGRATION RATES OF THE NATURALLY OCCURRING URANIUM ISOTOPES

Name	Mass	Abundance, per cent	Disintegrations per sec per gm U	Half-life, years
UI	238	99.274 [5]	$1.223 \times 10^4$	$4.51 \times 10^9$ [5]
AcU	235	0.71 [5]	$0.055 \times 10^4$	$7.07 \times 10^8$ [5]
UII	234	0.00518 [5]	$1.223 \times 10^4$	$2.35 \times 10^5$ [5]
Total		100	$25.01 \times 10^4$ [4]	

A typical alpha-particle ionization chamber that may be used for obtaining absolute measurements is shown in Fig. 100. The samples, usually deposited in a thin layer or electropolated on platinum, are placed on electrode A. The electric field between the electrodes is of the order of 1,000 volts per cm. All alpha particles that cross the sensitive volume of the chamber, and produce in the order of 3,000 ion pairs, give rise to a voltage pulse on the collecting electrode which may be amplified with a linear amplifier and counted by a conventional scaling circuit. Usually about 50 per cent of all the alpha particles emitted by the sample travel through the sensitive volume, and each of these will cause a count in the instrument. Most of the remaining 50 per cent will go into the electrode and be absorbed there. However, a small fraction of these particles (approximately 2 per cent for 5-mev alpha particles) will be reflected by the electrode and counted in the chamber, as indicated by the measurements of Ghiorso *et al.* [6] on reflection of alpha particles by platinum.

Alpha-particle ionization chambers calibrated by a primary standard can be used for direct determination of absolute disintegration rates of other alpha emitters, which was the technique used by Kohman for the above-mentioned determination of the rate of emission of alpha particles from  $\text{Ra}^{226}$ . The probable error of such absolute determinations of alpha disintegration rates may be of the order of 0.5 per cent. In measurements of aged alpha-particle samples it is necessary in some instances to take into account diffusion of the radioactive material into the sample support [39].

**13.4. Standards for Beta-particle and Gamma-ray Emitters.** Technically, standardization of beta-particle and gamma-ray emitters presents a much more difficult problem than that of alpha particles. The energy distribution of beta particles as well as the scattering of these particles is responsible for much of the difficulty. In the case of the gamma rays, the problem arises partly from the fact that these radiations have been detected mainly through their secondary particles. Several independent methods of standardization are available. The most important of these are as follows:

1. Standardization by coincidence measurements.
2. Long-life beta-particle standards.
3. Beta-particle standardization by direct measurement of the charge of the particles.
4. Indirect standardization by calorimetric measurement of the rate of energy emission.
5. Ionization measurement of beta particles and gamma rays.
6. Standardization of beta-counter geometry.

A discussion of each of these methods is given below. It should be borne in mind that the response of all known instruments to beta and gamma rays is a function of the energy of these radiations. The standardization of the

emission and disintegration rates of different radioactive isotopes will therefore depend to a considerable extent on the decay scheme of each isotope. When one radioactive isotope has been satisfactorily standardized, generally it cannot be used to serve as a standard for measurement of other radioactive isotopes with different level schemes and different beta-particle and gamma-ray energies.

**13.5. Standardization by Coincidence Measurements.** By means of an electronic circuit first developed by Rossi [7], it is possible to record simultaneous occurrence of pulses generated in Geiger counters with a resolution of  $10^{-7}$  sec. Coincidence counting was originally used as one method for the study of the level schemes of radioactive isotopes. However, in radioactive isotopes where the level scheme is completely known, this method may be used to determine the absolute disintegration rate of such isotopes as well as the efficiency of the beta and gamma counters. A review of the early literature is given by Dunworth [9]; more recently various investigators used the coincidence method for standardization, for example, M. Wiedenbeck [10].

A number of radioactive substances emit beta particles that are followed in less than  $10^{-7}$  sec by one or more gamma-ray quanta. In some radioactive substances the direction taken by the beta particle with respect to the gamma ray that follows is isotropic; in others, as shown by Brady and Deutsch [8], there may be a slight dependence on the angle. For the purpose of coincidence standardization, however, the distribution should be isotropic.

To derive the simple relationships necessary for this method, consider a radioactive isotope emitting a single beta particle followed by a single gamma ray. Denote the disintegration rate of the radioactive isotope by  $R$ , and assume a beta counter counts at the rate  $B$ , a gamma counter at rate  $G$ , and a coincidence device records beta-gamma coincidences at the rate  $C$ . Let us further assume that the over-all efficiency of a beta counter is  $e_\beta$  and that of the gamma counter  $e_\gamma$ . For the moment neglect the various corrections, background, etc., and write the self-evident expressions for  $B$ ,  $G$ , and  $C$  as follows

$$Be_\beta = R \quad G e_\gamma = R \quad \text{and} \quad C = R e_\gamma e_\beta$$

These relationships assume that there is no correlation between the direction of each beta particle and the following gamma quantum and that only one gamma ray follows the beta particle and is not internally converted. Solving for  $e_\beta$ ,  $e_\gamma$ , and  $R$ ,

$$e_\beta = \frac{C}{G} \quad e_\gamma = \frac{C}{B} \quad R = \frac{BG}{C}$$

In an actual experiment the quantities  $B$ ,  $G$ , and  $C$  are obtained by taking into

account a number of corrections. Without going into detail these are as follows: (1) Subtract the background from the single and coincidence counting rates measured. The coincidence background is due mainly to cosmic-ray particles which pass through both counters. (2) Subtract the counting rate caused by the gamma rays in the beta counter. The beta particles usually do not get into the gamma counter. (3) Correct all counting rates for finite

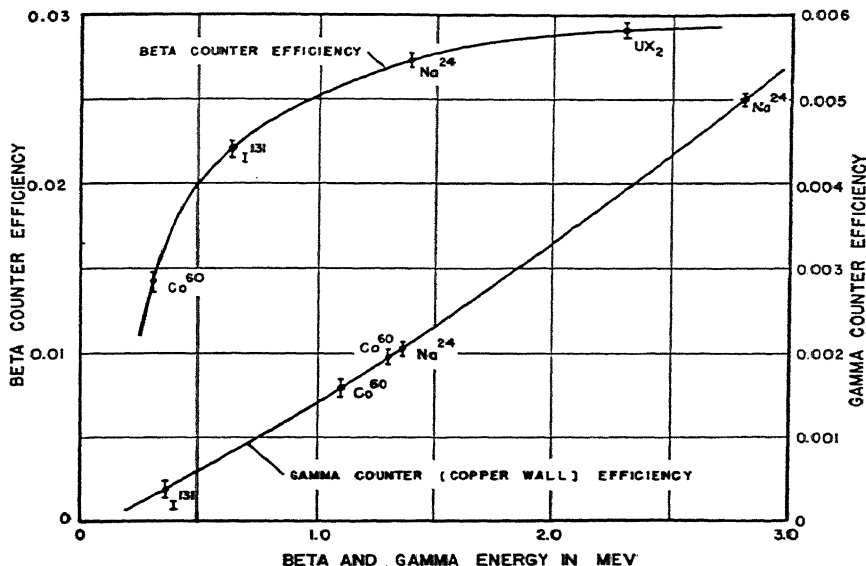


FIG. 101. Typical counter efficiency curves obtained with a  $\text{UX}_2$  standard and by the coincidence method. The curves indicate the over-all efficiencies for the particular counters and geometry used in the measurements; consequently, the curves are influenced by geometry, scattering, and absorption as well as by the absolute efficiencies of the counters for responding to each incident particle or photon. [Unpublished data of H. Anger.]

resolving time of each counter. (4) Correct the counting rates  $C$  for accidental coincidences between beta and gamma counts. (5) Finally, evaluate the statistical errors by the method of propagation of errors. All these corrections will influence the over-all accuracy of measurement. Primary standardization requires prolonged counting in order to reduce errors due to statistical fluctuations. The best available determinations of absolute disintegration rates have about 1 per cent probable error. The technique of coincidence counting measurements is rapidly being improved [11-13]. The use of fluorescent counters appears to increase the sensitivity and

accuracy of the method [14] by at least a factor of 10. Among the radioactive isotopes that have been standardized by this method are  $\text{Co}^{60}$ ,  $\text{Na}^{24}$ ,  $\text{Au}^{198}$ ,  $\text{I}^{128}$ ,  $\text{I}^{131}$ , and  $\text{Fe}^{59}$ . Other isotopes can be used later when their level schemes are better known.

When the beta- and gamma-counter efficiencies obtained by this method are plotted as a function of the energy of the beta particles or gamma rays, curves such as those shown in Fig. 101 are obtained. In this diagram the strong dependence of counting efficiency on energy may be clearly seen. If one wishes now to standardize other radioactive isotopes that have no convenient level scheme for coincidence measurement, it is possible to interpolate on the beta- and gamma-counter efficiency curve and obtain the efficiency for any other energy, provided that the geometry is not changed. In this way counter efficiencies for pure beta emitters, such as  $\text{P}^{32}$ ,  $\text{C}^{14}$ , and  $\text{S}^{35}$ , may be determined [42]. The efficiency curve for counters may be predicted by some semiempirical considerations. Although it seems to be entirely permissible to extrapolate on the gamma-counter efficiency curve for energy regions where selective K or L absorption in the counter is unimportant, this is not quite so with the beta-counter efficiency curves because the energy distributions of beta particles from different radioactive isotopes do not follow the same law. The shape of the beta spectrum depends on whether or not the spectrum is allowed or forbidden and there is not sufficient experimental and theoretical evidence to make very accurate predictions of the shape of the spectrum. In spite of this, however, no great error is incurred in most cases when the extrapolation technique is used. Similar difficulties arise when positron emitters are to be standardized. Here the shape of the low-energy end of the positron spectrum is very uncertain and the best method appears to be the use of the annihilation radiation (0.51-mev gamma rays). The efficiency for counting annihilation gamma rays may be easily determined from the gamma-counter efficiency curve.

**13.6. Long-life Beta Standards.** It is useful to compare the results of disintegration-rate determinations by the coincidence method with other independent methods. Chronologically, the first such method was one using gravimetrically prepared samples. In addition to primary standards, the use of long-life secondary beta-particle and gamma-ray standards is very important in routine measurements because their periodic measurement informs one of the changes of sensitivity of counting and ionization measuring instruments.

*a. Uranium Standard.* Fermi was one of the first to use radiations from  $\text{UX}_2$  as beta-particle standards. The usual practice is to prepare a pure sample of  $\text{U}^{238}$  and then wait long enough for radioactive equilibrium to be reached between  $\text{UI}$ ,  $\text{UX}_1$ ,  $\text{UX}_2$ , and  $\text{UZ}$ . As is well known in a case of radioactive equilibrium, the rate of disintegration of the parent isotope is



the same as that of the daughter isotopes. Thus in accordance with the scheme given in Chap. 7 (uranium-radium series), the rate of  $UX_2$  betas emitted is very nearly the same as the rate of UI alpha particles per unit time. For the actual values, see Table 35. In practice the prepared uranium sample is carefully weighed and its rate of disintegration by alpha emission accurately measured. From these data the rate of  $UX_2$  beta emission is found directly. The level scheme of  $UX_2$  gives rise to some complication because the beta spectrum is not simple. A recent level scheme proposed by Bradt and Scherrer [38] is shown in Fig. 102. The beta radiations of  $UX_2$  represent 99.85 per cent of all disintegrations, the rest, 0.15 per cent, being gamma rays which produce UZ. Furthermore, 98 per cent of  $UX_2$  beta particles is of 2.32 mev maximum energy, while 2 per cent consists of one or more beta particles of 1.5 mev in coincidence with gamma rays. If a known quantity of uranium is deposited on a foil and allowed to equilibrate with  $UX_1$  and  $UX_2$ , a combined alpha-beta standard source can be obtained. The alpha particles of UI, UIII, and AcU and the beta particles of  $UX_1$  have to be absorbed if the  $UX_2$  beta emission is to be used for standardization purposes. This is readily done with 25 mg per  $cm^2$  aluminum foil which absorbs all alpha and  $UX_1$  beta particles but only about 5 per cent of the  $UX_2$  beta particles.

A uniform deposit of uranium on foil may be prepared with a colloidal suspension of uranium oxide. Pure uranium oxide is ground in a quartz ball mill for 12 hr. The material is taken up by a suspending agent such as chloroform or ethyl alcohol and centrifuged to precipitate the coarse particles. The remaining colloidal suspension is then evaporated in a standard holder by the radiant heat from a General Electric heat lamp. The amount of material deposited is determined by weighing the aluminum foil before and after deposition. Some of the uranium standards are still inconvenient when they are used with Geiger counters because there may be appreciable scattering of the beta rays in the bulk of the standard sample itself. If the mass of the sample is reduced so that this effect is negligible, the emission rate of beta particles also decreases to only a few times that of the background, thus making the determinations less accurate. These points have been noted by Broido *et al.* [15], who made use of methods for quantitative separation of  $UX_1$  and  $UX_2$  from aged uranium solutions, using lanthanum carrier. One of the methods used is as follows:

To 25 ml of an aqueous solution containing 0.1 gm of uranium per milliliter add 25mg of lanthanum carrier and 1.5 ml of 27*N* hydrofluoric acid. Centrifuge and wash the resulting lanthanum fluoride and  $UX_1$  precipitate with 10 ml of 6*N* hydrofluoric acid. Collect the supernatants and carry them through a second lanthanum fluoride precipitation to check for losses. Dissolve each precipitate in a mixture of 8 parts nitric acid and 1 part boric



absorber of 10 mg per cm<sup>2</sup>. Usually the RaD is electrolytically deposited in a very thin surface layer, and if sufficient time is allowed, the alpha disintegration of polonium may be measured to verify the sample activity. RaE has a single beta particle and no gamma rays, but the beta transition is classified as forbidden so that the spectrum is different from that of beta particles in allowed transitions. There is also one alpha particle emitted for about every million betas. The Bureau of Standards [16] has made a

TABLE 36. RADIATIONS FROM RAD, RA E, AND PO

Element	Alpha particle, mev	Beta particle, mev	Gamma ray, mev	Half-Life
RaD 210	.....	0.025	0.047	22 years
RaE 210	4.87(10 <sup>-4</sup> -10 <sup>-5</sup> %)	1.17	.....	5.0 days
Po 210	5.298 (100%)	....	0.8 (weak)	140 days

number of standards of this type for distribution among radioisotope workers. These standards can be used to calibrate the approximate efficiency of Geiger counters for beta radiations. However, when this is done a number of rather complex factors have to be considered. These will be discussed in Sec. 13.10.

In the method given by Broido *et al.* [15] for preparation of RaE standards, chemical separation of RaE from RaD and Po<sup>210</sup> is utilized. A mixture of RaD, E, and F should be taken up in 10 ml of 0.1*N* hydrochloric acid. Add 20 mg powdered nickel, heat the mixture to 80°C, and stir for 3 min. The powdered nickel then takes up the RaE and F. Centrifuge and wash the precipitated nickel three times with 0.1*N* hydrochloric acid. Dissolve the nickel with ~ 1 ml aqua regia and dilute to 10 ml. The nickel may now be separated by addition of 20 mg bismuth carrier and then an excess of ammonium hydroxide. Bismuth hydroxide precipitates carrying RaE and fluorine but leaving the nickel in solution. In order to separate the RaF from the RaE an ion exchange column may be used. The bismuth hydroxide is dissolved in hydrochloric acid diluted to an acidity of 0.1*N* and passed through a 1-cm diameter column made up of 10 ml 40-60 mesh 1R-1 resin. The bismuth and RaF are absorbed in the column. The bismuth may be eluted with 3*N* nitric acid (approximately 10 ml.) leaving the RaF in the column. The purity of the RaE preparation may be checked by half-life and absorption measurements and with growth curves of the alpha activity of RaF (Po<sup>210</sup>). The absolute RaE disintegration rate may be obtained by measuring the absolute RaF activity,  $I_f(t)$ , in an alpha-particle chamber and extrapolating back to the amount of RaE at zero time  $I_e(0)$ , using the equation

$$I_e(0) = \frac{I_f(t)(1 - \lambda_e/\lambda_f)}{e^{-\lambda_e t} - e^{-\lambda_f t}}$$

where  $\lambda_e$  and  $\lambda_f$  are the decay constants of RaE and RaF, respectively. The beta disintegration rate of the RaE sample at time  $t$  is then

$$I_e(t) = I_e(0)e^{-\lambda_e t}$$

**13.7. Beta-particle Standardization by Direct Measurement of the Charge of the Particles.** If a beta emitter is placed on an electrically insulated support in the center of an evacuated metal chamber, the charge due to the beta particles may be observed when these particles are absorbed by the walls. This method has recently been used by Failla [19] and his associates to obtain standardization of some important beta-active isotopes, such as  $P^{32}$ . If precautions are followed to avoid secondary effects, the results obtained by this method are within 5 per cent of other independent methods. The value of this method lies in its independence from other methods. For secondary standardization its use would probably not be convenient for routine measurement.

**13.8. Indirect Standardization by Calorimetric Measurement of the Total Energy.** Zumwalt and his associates [20] employed an isothermal calorimeter for the determination of the rate of heat evolution from radioactive phosphorus when the beta particles were stopped in the calorimeter. The measurement was carried out with about 25 millicuries of  $P^{32}$  at the temperature of liquid nitrogen, using the rate of evaporation of nitrogen at constant pressure as the measure of the heat output. With measurements of this kind it is necessary to determine certain constants in order to evaluate the absolute disintegration rate. The total energy output is the product of the emission rate and the average energy of the beta particles; hence, it is essential to determine the average energy independently from measurements of the energy distribution of the  $P^{32}$  beta particles. A small fraction of the beta-particle energy is converted into *Bremsstrahlung* which may not be completely absorbed in the calorimeter. The *Bremsstrahlung* accounts for about 1.3 per cent of the total energy of  $P^{32}$ . A small fraction of the energy lost by ionization in the calorimeter also may be transformed to chemical energy which will not change the temperature of the device. The over-all probable error in calorimetric determinations of the total emission rate is between 2 and 3 per cent.

**13.9. Ionization Measurements of Beta and Gamma Rays.** Many investigators prefer to use ionization chambers for routine determinations of the disintegration rates of various radioactive isotopes. In x-ray dosimetry a number of types of ionization chambers have been in use for a long time, and for special problems in connection with radioactive isotopes highly sensitive chambers and electrometers have been developed (see Chap. 12).

The ionization produced by a charged particle traversing a gaseous medium is proportional to the energy loss of the particle to the medium. The factor of proportionality appears to be constant over wide limits of particle energy. Thus, ionization measurements become measurements of energy loss in the medium when the factor of proportionality is known. The energy loss per ion pair for beta particles in air is considered to be about 32.5 ev. In practice absolute determination of energy loss by ionization becomes a more complex problem because of the wall effects in the chamber. Both beta particles and gamma rays are scattered back from the walls of the chamber, thus increasing the ionization in the sensitive volume. Secondary electrons produced in the walls by gamma rays also contribute to the ionization, often to an extent that is difficult to estimate. Consequently, considerable caution is necessary in the interpretation of ionization phenomena. In practice, two different ionization-chamber constructions are possible in which these complex conditions may be accurately analyzed. These are chambers in which the wall effect is negligible and chambers in which the wall effect is made the most important contribution to the ionization (see also Sec. 12.2).

*a. Gas-wall Chambers.* These chambers are best suited to standardization of low-energy gamma rays and low-energy beta emitters such as  $H^3$  and  $C^{14}$ . Such instruments are usually constructed with a central electrode of fine wire and a concentric cylindrical electrode consisting of a coarse mesh made of fine wire. It is important that the solid walls of the chamber be placed farther from the outer electrode than the greatest range of the beta particles being measured or the range of the secondary electrons from the wall when gamma rays are measured. Ionization chambers of this kind often operate at higher than atmospheric pressure in order to reduce the linear dimensions required to meet this condition.

For an isotope in gaseous form, emitting beta particles of average energy  $\bar{E}_\beta$ , the disintegration rate  $R$  per unit gas volume may be expressed in terms of the measured ion current  $I$  by the simple formula

$$R = \frac{WI}{eV\bar{E}_\beta}$$

where  $e$  = electronic charge

$W$  = average energy required to form one ion pair

$V$  = volume of sensitive region

For x- and gamma rays the energy flux  $F$  may be determined from the expression

$$F = \frac{WI}{\mu_m \rho V e}$$

where  $\rho$  = density of chamber gas

$\mu_m$  = mass-absorption coefficient for gamma rays in the gas (sum of photoelectric and Compton absorption coefficients)

If the gamma rays are monochromatic, with energy  $E_\gamma$ , the flux of photons crossing the chamber is then  $f = F/E_\gamma$ . By taking into account the absorption and the solid angle subtended by the instrument with respect to the source, the emission rate of gamma rays from an external source can be determined.

*b. Solid-wall Ionization Chambers.* Gas-wall chambers are not easy to construct and operate, and their usefulness breaks down at energies higher than a few hundred kev. However, ionization chambers with solid walls may be used for x-rays and gamma rays from a few kev to very high energies. Most of the ionization chambers in routine use for radioisotope measurements and dosimetry are of the solid-wall type, a very popular form of which is the so-called "thimble" chamber. The methods for absolute measurement of gamma rays and energy flux with such instruments were elucidated by Gray [21]. To facilitate interpretation, the ionization chambers are constructed in such a way that, for practical purposes, nearly all ionization in the sensitive volume comes from secondaries initiated in the solid electrodes of the instrument while the number of secondaries produced in the gas add negligibly small ionization. A simple relationship holds true between the energy  $E_s$  absorbed per unit mass of the solid medium of the wall and the ionization  $I$  produced in a small air-filled cavity of that medium, if the dimensions of the cavity are small compared to the mean distance traveled in the air by the secondary electrons and if the dimensions of the solid are large compared to the distance traveled by the secondary electrons in the solid. This relationship, sometimes also called the Bragg-Gray principle, may be written as

$$\frac{IW}{eV\rho} = \frac{E_s}{S}$$

The meanings of  $W$ ,  $V$ ,  $\rho$ , and  $e$  are the same as above, while  $S$  is the relative stopping power per unit mass of the solid medium versus air. With the knowledge of the absorption coefficient  $\mu_m$  of the solid medium of the ionization chamber the gamma-ray energy flux  $F$  at some point near the center of the chamber is then

$$F = \frac{E_s}{\mu_m} = \frac{IWS}{\mu_m V e \rho}$$

Since gamma rays of a few hundred kev do not lose much energy traversing a few feet of air or through the walls of small ionization chambers, the rate of emission from a point source of monoenergetic gamma rays of energy  $E_\gamma$

at a distance  $r$  from a suitable ionization chamber is approximately

$$R = \frac{4\pi r^2 I W S}{e \rho \mu_m V E_\gamma}$$

Unfortunately, in precision measurements some corrections are again needed. Among these an important one is that due to Compton scattering. Uncertainties come in also in the measurements of various quantities in the above expression, notably in  $I$ ,  $W$ ,  $\mu_m$ , and even  $V$ , while  $E_\gamma$  and  $S$  may be measured quite accurately. In measurement of  $I$ , care must be taken that all ion pairs formed in the sensitive volume are collected without allowing recombination or multiplication. This is best accomplished by the use of an inert gas or dry air and with conveniently shaped ionization chambers which have a well-defined plateau. As long as the collecting potential is held within the limits of the plateau, 100 per cent of the ions are collected. The biggest uncertainty at the present time exists in the value of  $W$ , the energy absorbed to form one ion pair, as indicated by the fact that independent methods and investigators have arrived at rather widely differing values for  $W$ . One of the best ways to measure this value is by counting the number of droplets produced in cloud-chamber tracks of electrons. The probable error of about 1 per cent for the value quoted above places a limit on the absolute accuracy of gamma-ray disintegration evaluations and also on estimates of the energy absorbed in matter for one roentgen of gamma rays and x-rays. Assuming this value of  $W$  to be correct, the conversion factor is  $84 \pm 1$  ergs per gm in air.

Notwithstanding the experimental difficulties several excellent determinations were made of the ionization of weighed amounts of radioisotopes, especially that of radium. One gram of radium, when shielded with  $\frac{1}{2}$  mm of platinum gives, at 1 meter distance, ionization in air equivalent to  $0.84 \pm 0.01$  rhm [22 to 25]. Knowing the absorption coefficients, the formula above may be used to calculate the expected energy absorption or the ionization from any radioisotope even with more than one gamma ray present. Marinelli *et al.* [26] have recently published such data, some of which are reprinted in Table 37. The case of radium is more complex than most other radioactive isotopes because when in equilibrium with its disintegration products a Ra sample emits some 16 gamma rays with different intensities and energies. Nevertheless the theoretically calculated value agrees quite well with the experimental data [27].

The measuring methods for ionization and energy absorption are constantly being refined. One particularly suitable arrangement for the study of the wall effect and scattering in ionization chambers is that constructed by Failla [28]. This ionization chamber has parallel walls that form the electrodes. The gas pressure, the distance of the walls, their material and





readily obtained. However, since the method is not entirely foolproof, it may be useful to consider some of the factors that influence the counting rate, and some of the methods whereby simple and fairly reliable standardization may be achieved for almost any radioactive isotope.

Theoretical considerations as well as actual tests carried out with coincidence counters make it fairly certain that each beta particle that passes through the sensitive volume of a counter will be counted. The gas pressure in most counters is made high enough to make certain that each beta particle will make at least one ion pair. Similarly, a gamma ray will be counted if at least one of its secondary particles passes through the sensitive volume.

It is generally assumed that the sensitive volume is a well-defined space within the counter. However, this is not strictly true. Measurements of counter efficiency indicate that the apparent sensitive volume changes with beta-particle energy in bell-jar counters [40]. Furthermore, these counters are less sensitive in the region near the end by the window and in the region close to the surface of the cathode (cylindrical electrode) than they are near the anode (wire electrode). In order to avoid using the insensitive regions of the counter a circular aperture about one-half the diameter of the counter is placed in front of the counter window, as shown in Fig. 103. All beta particles within the cone subtended by the aperture from a point source are counted, provided that they have enough energy, penetrate the counter window, and reach the sensitive volume. The aperture may be made in a piece of brass plate thick enough to stop all beta particles. From Fig. 103 it is seen that the solid angle subtended by the aperture at the source is

$$\Phi = 0.5(1 - \cos \alpha)$$

$$\text{where } \tan \alpha = \frac{b}{a}$$

The value of  $\Phi$  is more generally referred to as the geometrical efficiency.

In performing measurements on beta particles it is important to keep in mind the effects of multiple scattering. The importance of scattering on beta particles may be emphasized by observing the tracks left by these particles in a cloud chamber. They are observed to be deflected many times, often through large angles. A complete study of the scattering process is not available, but it is known that the beta particles are scattered both by nuclei (Rutherford scattering) and by the atomic electrons. Sometimes the scattering is inelastic, giving rise to x-rays (*Bremsstrahlung*). The scattering and absorption phenomena are usually superimposed and together modify the distribution and intensity of beta particles.

The above "geometrical-efficiency" formula would be correct if the sample were a point source and of negligible mass and if there were no air, window and self-absorption, or scattering by supports. Since these conditions cannot

be achieved in practice, it is necessary to take them into consideration and make detailed corrections for each factor. Most of the corrections have empirical bases because it would be difficult to make exact calculations of the combined absorption and scattering effects.

When absorbers are placed between counter and source and the counting rates are plotted against absorber thickness, the well-known absorption curve is obtained. Near the end of this curve at large absorption thicknesses, the logarithm of the number of transmitted beta particles is, in many instances,

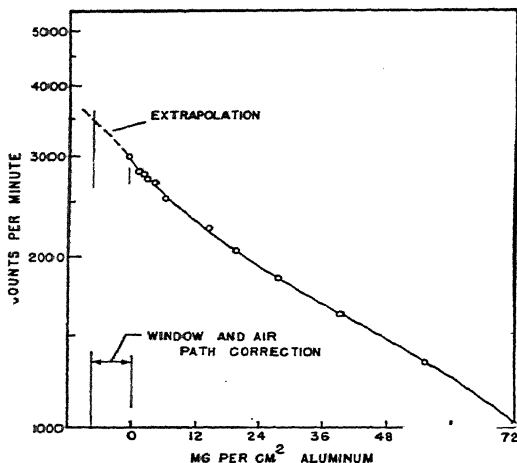


FIG. 104. Typical aluminum absorption curve for RaE. The units of abscissa represent only the thickness of aluminum absorber. To this must be added the equivalent aluminum thickness of the counter window and of the mean path of the particles in air. The extrapolation shown is subject to considerable uncertainty. [Redrawn from L. R. Zumwalt, U.S. Atomic Energy Commission Report MDDC-1346, 1947.]

proportional to the thickness. At very small absorber thicknesses this relationship does not hold. In fact, theoretically one would expect the absorption coefficient to be zero at very small absorber thickness because of the shape of the Fermi distribution of beta-particle energies. A typical absorption curve is shown in Fig. 104. Scattering plays an important role in the shape of the absorption curves. Hence there is no well-defined "accepted" absorption curve for any radioactive isotope since the curves vary markedly with the experimental arrangement. It is customary to use extrapolation procedures for correcting the effect of window absorption on the observed beta-particle counting rates [16]. Starting with the counter window as the only absorber additional aluminum absorbers are added in steps; then assuming that the absorption curve is logarithmic, it is extra-

polated back to zero thickness. Actually such a procedure is only permissible if the extrapolation is very small. Using soft beta emitters, for example, the absorption curve with the absorber close to the counter window is usually very different from that with the absorber right over the sample [41].

The angular distribution of beta particles from a point source of small mass is isotropic. A sample support of finite thickness, or an extended

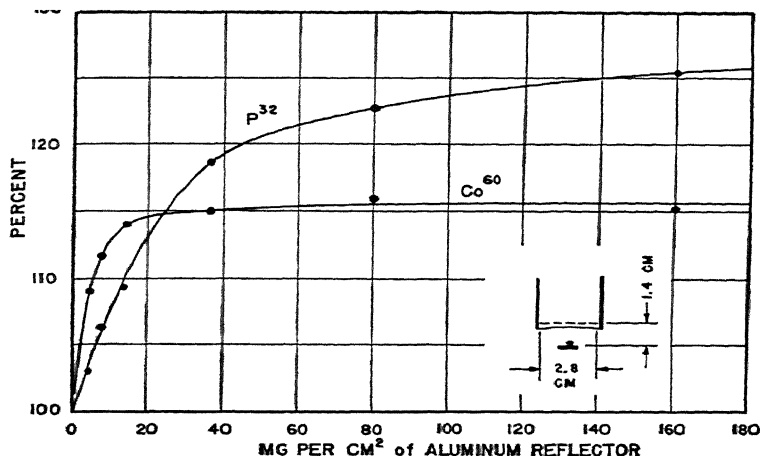


FIG. 105. Backscattering of  $P^{32}$  and  $Co^{60}$  beta particles ( $E_{max} = 1.72$  and  $0.3$  mev, respectively) as a function of sample support thickness. The counting rate is given in per cent of the rate obtained with a sample support of negligible thickness. The samples were point sources, and the counting geometry is indicated in the diagram showing the position of sample, counter window and sensitive region of counter (broken line). [Unpublished data of H. Anger.]

source with finite thickness, changes the isotropic distribution into one preferring a direction perpendicular to the plane of the sample. The first effect is usually called backscattering. The magnitude of this depends on the beta-particle spectrum and the atomic number and thickness of the sample support. Using an aluminum sample support, the effect of different thicknesses on the counting rate of  $Co^{60}$  and  $P^{32}$  is plotted on Fig. 105. It is apparent that if the support thickness is greater than one-half the amount of absorber necessary to stop all beta particles the counting rate does not increase with support thickness. This condition is often called "saturation backscattering." The dependence of saturation backscattering on atomic number of sample support is shown in Figs. 106 and 107. Some beta-particle standards are mounted on supports that give saturation backscatter-

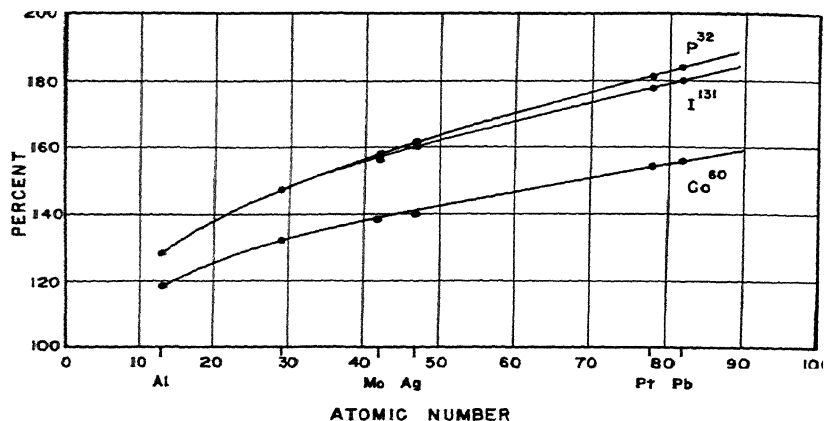


FIG. 106. Saturation backscattering as a function of the atomic number of the backscatterer. The ordinate represents per cent of the counting rate obtained with a sample support of negligible thickness. [Unpublished data of H. Anger.]

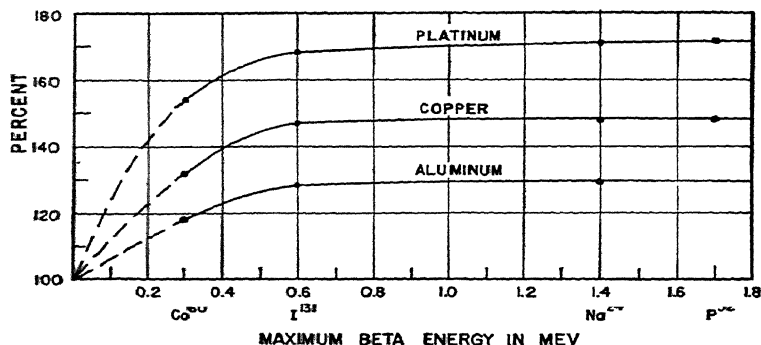


FIG. 107. Saturation backscattering from aluminum, copper, and platinum for a point source as a function of maximum beta-particle energy. The ordinate represents the counting rate with respect to a source with no backscatterer (negligibly thick supporting film). [Unpublished data of H. Anger.]

ing, and when these are compared with various beta-particle samples, the variations in backscattering must be taken into account. Absorption curves taken from samples with saturation backscattering are different from those taken with very thin sample supports because the scattered particles increase the relative number of soft beta particles entering the counter.

Backscattering can be very nearly eliminated by using extremely thin

sample supports consisting of films of materials, such as nylon, polyethylene, or aluminum, which have low atomic weights. Scattering by the intervening air and parts of the apparatus can, of course, be greatly reduced by proper design and by placing the entire unit in an evacuated box, as shown in Fig. 103.

Routine counting of samples usually necessitates deposition of several milligrams of active material on some kind of standardized sample holder, as described in Chap. 17. For a thin sample the computed and experimental

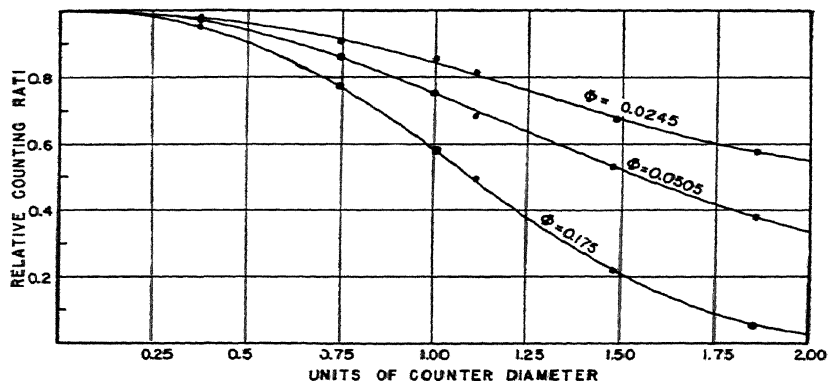


FIG. 108. Relative counting efficiency (for  $P^{22}$ ) as a function of the lateral displacement of the source from the axis of the counter.  $\Phi$  denotes the geometric efficiency when the sample is on the counter axis. [Unpublished data of H. Anger.]

efficiency decreases as a function of the distance the sample is displaced laterally from the axis of the counter in a manner indicated in Fig. 108. Samples with finite thickness introduce an additional effect, a combination of absorption and scattering in the sample material, often called self-absorption. Along with this there is also distortion of the isotropic angular distribution of the beta particles. A counter, however, may be calibrated for measuring samples of any thickness by obtaining first a standard self-absorption curve for a given substance. This may be done in two different ways. A dilution curve is obtained if a sample of known radioactivity and negligible mass is progressively diluted by addition of stable carrier. With samples of identical intensity and sample area the counting rate decreases progressively as the mass of carrier is increased. Another method of measuring self-absorption is by the use of radioactive samples with the same specific activity but different total mass or thickness. Instead of a linear relationship between sample thickness and counting rate, one obtains a typical saturation curve

reproduced for  $C^{14}$  in Fig. 109. When samples of finite thickness are used, nonuniformity of thickness results in a decreased counting rate as a consequence of variable self-absorption. The precise methods for obtaining calibrations for self-absorption have been discussed by Yankwich *et al.* [31].

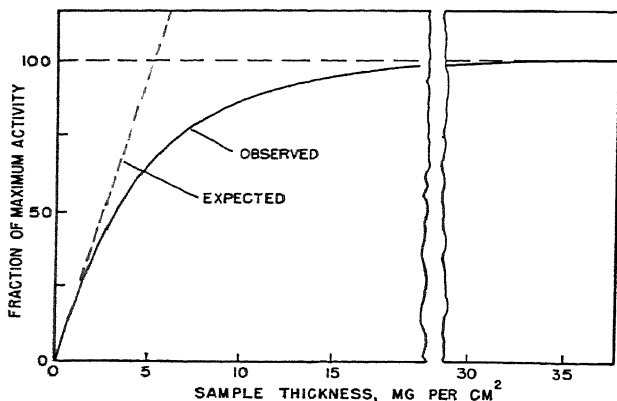


FIG. 109. Fraction of maximum observable activity of  $C^{14}$  in a  $BaCO_3$  sample as a function of sample thickness. The broken line indicates the activity which would be observed in the absence of self-absorption. [Redrawn from P. E. Yankwich, T. H. Norris, and J. Huston, *Anal. Chem.*, **19**, 439 (1947).]

**13.11. Secondary Beta-particle and Gamma-ray Standards.** The Bureau of Standards has made available a number of secondary standards. Among these are gamma-ray standards of Ra and of  $Co^{60}$  (the latter standardized by the coincidence method) and RaE beta-particle standards. In the near future secondary beta-particle standards of  $Co^{60}$ ,  $I^{131}$ ,  $P^{32}$ , and  $C^{14}$  will also be added. In the meanwhile further work is going on in the search for a convenient long-lived beta standard with higher maximum energy than  $Co^{60}$ . Some laboratories and commercial enterprises have also prepared  $UX_2$  beta-particle standards.

**13.12. Standardization of Neutrons and Protons.** Detailed processes for standardization of neutrons are beyond the scope of this chapter, although some of the more important considerations of the problems that arise can be briefly stated. The number of fast neutrons is usually determined by measuring the radioactivity induced in some substance for which the interaction cross section is known accurately. From the point of view of biology, the ionization produced by fast neutrons is more important than the number of such neutrons. For some purposes, therefore, the ionization is often measured by a thimble ionization chamber. As in the case of gamma-ray

measurements, it is necessary to achieve the condition in which the ionization in the gas of the chamber is in equilibrium with the ionization within the chamber walls [21.32]. The neutrons exert their biological effects by the ionization produced by the secondary recoil nuclei; hence, it is important that equilibrium is also reached between the primary neutrons and the secondary protons in the material studied. Using 10 mev neutrons, equilibrium may be reached in about  $\frac{1}{8}$  in. of tissue, whereas with 100-mev neutrons the equilibrium is established only after the neutron beam has crossed some 3 in. of tissue.

The measurement of thermal neutrons may be done in several different ways. Among these is the activation of certain elements, e.g., indium, gold, or manganese, which have large thermal-capture cross sections. The neutron flux may be calculated from the observed disintegration rates of the isotopes produced and from the cross sections for the processes. An independent method utilizes neutron-induced fission of uranium measured with an ionization chamber. Each time the fission occurs a large burst of ionization appears in the chamber due to fission recoils, and the number of pulses, together with the cross section for fission, yields the absolute number of primary neutrons. If a neutron beam has thermal as well as fast components, separation of the components is possible to some extent by measuring the difference in radioactivity induced in detecting foils with and without cadmium shielding. The radioactivity induced in the foil shielded with a strong thermal neutron absorber is due only to faster neutrons.

Recently protons, deuterons, and alpha particles accelerated in cyclotrons have also been used in biological research. The measurement of a number of particles in such beams may be accomplished by two methods: (1) measurement of a total charge carried by the beam by means of a Faraday cage, and (2) by measurement of the ionization produced in air by a fraction of a monoenergetic beam at a given energy. From the known charge carried by each particle and the measured ionization, the number of particles in the beam may be calculated [35,37].

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## CHAPTER 14

### THE RADIOAUTOGRAPH

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**14.1. Introduction.** Established methods for analyses of radioactivity in biological specimens in which the activity of a suitable aliquot of treated tissue is determined will give valuable information concerning gross distribution of the material under study. However, the local distribution of active material in particular parts of an organ and in special cells of a tissue can be studied most effectively by the technique of radioautography. This technique, as the name implies, provides a self-photograph of a radioactive substance as a result of the action of alpha or beta particles on a photographic emulsion (gamma radiation will not be considered since it is of limited importance).

The usual photograph, taken with a camera, is the result of activation of silver bromide grains by photons; the energy state of the silver bromide is changed so that at some later time the silver may be easily reduced. Presumably, the action of alpha and beta particles is qualitatively similar to that of photons but quantitatively different because of the physical characteristics of the radiations. The alpha particle has a mass about 7,400 times that of the beta particle, and when both particles are of equal energy the beta particle will travel at a much greater speed than the alpha particle. When the alpha particle passes through an emulsion containing silver bromide grains, it proceeds in a straight line giving up energy to the grains along its path until it is brought to rest and neutralized. A radioautograph produced from an alpha source will show distinct tracks of closely spaced grains. The tracks, each produced by a single alpha particle, proceed in a straight line, lie in random directions, and end abruptly. In a picture from a beta source the situation is somewhat different, for in this case the velocity of the beta particle compared with that of an alpha particle of equal energy is much greater; the grains affected by a single particle are more widely spaced since the beta particle passes many grains too rapidly to induce an image on them. Furthermore, because of the small mass of the beta particle, it is easily deflected by atoms in its path and will not travel in a straight line. From this discussion, it will be recognized that with a beta source the picture obtained will consist of a random distribution of developed grains, no two of which can be ascribed to any one beta particle. In addition, the length of path of the beta particle through material is much greater than that of an

alpha particle since the former loses energy more slowly. As a result, an image produced by a point source of a beta emitter will be quite large; for example, from a point source of beta particles with an average path length of  $2,000\ \mu$ , a symmetrical image approximately 4 mm in diameter is produced.

These circumstances are unfortunate for they limit the resolution of beta autographs and decrease the possibility of determining with certainty the origin of beta radiation from tissue samples. It is especially unfortunate since many of the isotopes most useful in biological studies, such as carbon, sulfur, sodium, and iodine, are beta emitters.

The alpha emitters are heavy metals, and their present importance falls primarily in the realm of health protection of workers in atomic-energy plants and in the effects of the atomic bomb on plant and animal life.

**14.2. Techniques for Preparing Radioautographs.** A radioautograph of a large sample of tissues containing radioactive material, such as half a kidney or a smooth bone surface, is relatively simple to produce. The block of tissue is held in firm contact with the film for the desired length of time, and the film is then developed, after which, areas of blackening are easily correlated with regions of the tissue block. The problem of more accurate localization to particular structures in the tissue is more complex. In this type of radioautograph the following factors are of particular importance: the resolution or separation of darkened areas and their restriction to an area the same as that of the source should be as great as possible in order that localization may be accurate; the sensitivity must be adequate to detect even small amounts of radioactive material; and, for the purpose of comparing concentrations of material in various areas, it is necessary that the range of response be as great as possible. These factors may be controlled, in so far as they are independent of the type of radiation, by manipulation of the various conditions discussed below.

The most important aspects to be considered are contact between emulsion and specimen during exposure and alignment of radioautograph and specimen after development. Contact and alignment may be secured (1) by mounting the section and emulsion on separate supports such as microscope slides, (2) by spreading liquid emulsion or stripping film over the section, or (3) by mounting the section on the emulsion.

In the first method (1) the tissue is prepared by the usual histological techniques, and one or more sections are mounted on a microscope slide or cover slip. Factors such as solubility of the radioactive material and section thickness, discussed in detail below, must be carefully controlled for best results. The slide holding the unstained section is then placed over the proper type of film, section and emulsion together, and firm, even pressure is applied for the duration of the exposure. After developing the film and staining the section the two may be easily compared for evidence of gross

distribution of active material, but microscopic examination is more difficult. A frame designed to fix the slide or cover slip with respect to the film is of some value for this purpose. The film and slide are fixed in their respective frames and held together by alignment pins or angles during exposure. After separate development and staining they may again be aligned and the tissue slide fixed to the film in the original position. Both components may then be removed from their frames. This procedure will yield a fairly satisfactory preparation for microscopic examination under low power and, depending on the thickness of the tissue preparation, perhaps under high power. The use of the oil-immersion objective, which is necessary for cellular localization of the radiation source, is impossible with this type of preparation. A microscope comparator may be used whereby tissue section and film may be aligned by a reference mark, such as a spot shadow of a small piece of gold leaf, and picture and tissue examined simultaneously. Alignment in this instance is not accurate enough to yield definite information concerning cellular origin of radiation.

The problems of realignment are solved by either of the last two methods (2 or 3) listed above. In the second method, the section is placed on a slide or cover slip, stained or not depending on the radioactive material being studied, and covered with emulsion. If the section is stained prior to exposure, it may be held on the slide by a very thin coating of collodion. Liquid emulsion, prepared by formulas given in the literature or melted from a plate or film, is then spread over the tissue. Preparation and handling of liquid emulsions is an art, and great care must be taken to avoid excessive grain size and high background fog. In addition, it is often difficult to obtain a uniform and thin film necessary for preparation of good radioautographs. However, a more satisfactory procedure is the use of an emulsion obtainable in the form of stripping film, which is prepared so that the emulsion may be removed from its backing and cemented over the section. After the proper exposure and development of the film, the section is stained if this has not already been done. Here and in the procedure outlined below, the usual histological stains are of limited use because of the strong affinity of the gelatin of the emulsion for the dyes. A completely satisfactory staining technique that gives excellent tissue differentiation and does not stain the emulsion has not yet been described. Another drawback in using liquid emulsion or stripping film is distortion of the emulsion through handling. This distortion is, of course, most serious in the case of liquid emulsions but also occurs to some extent with the stripping film, which frequently during the course of developing and staining becomes detached from the supporting slide. Although separation may be unavoidable, the radioautograph is still of value if developing and staining are finished carefully and the film with section attached is dried between blotting papers.

The last method (3) to be discussed eliminates the possibility of film distortion due to handling. Here, the cut sections are spread on water and (in the darkroom) are floated onto the emulsion supported on a microscope slide or other film backing cut to convenient size. The preparation is then air dried and stored in a lighttight container until the desired exposure time has elapsed. The emulsion acts as a fixative to hold the section to the slide. The collodion or paraffin is then removed, the picture developed, and the tissue section stained. The resultant preparation consists of the stained section lying on the emulsion. Tissue and radioautograph when examined microscopically under low magnification will appear to lie in the same plane. Under high magnification, they may be examined separately, since the tissue will appear at the uppermost level of focusing while the radioautograph will be brought into focus at a lower level.

Whatever method is used for assuring contact and alignment, several additional conditions should be examined and regulated to give the most effective results. During preparation of the tissues for sectioning, and through the staining procedure when the radioautograph is to be made after staining, it is important that the radioactive material not be dissolved from the tissue by any of the reagents used. For instance,  $I^{131}$  in organic molecules may be treated in the usual way without loss of material, whereas  $P^{32}$  is lost rapidly in acid fixatives. In the case of tissues containing the latter isotope, all solutions used in histological preparation prior to exposure for radioautography must be neutral. The question of solubility of radioactive material in the histological reagents may be answered only by checking all such reagents used prior to exposure of the film to the section. If the half-life of the radioactive substance under study is short, as with  $Na^{24}$ , or if the half-life is longer and the amount of material is small, it will be essential to reduce the time of histological preparation. By using small pieces, gentle heat, and agitation during fixing and embedding, the time required for preparing the tissue may be decreased markedly. The time may also be shortened and the problem of solubility eliminated as well by using frozen sections without fixing, although this introduces certain difficulties with regard to both histology and radioautography.

The thickness of the tissue sections used will influence the resolution greatly. Sections of 5 and 10  $\mu$  are preferable, since resolution is inversely proportional to section thickness provided that the latter is less than a limiting value, depending on the type of radiation. If the path length of the radiation is short, thickness of section is not critical with regard to resolution since particles from within the tissue will be absorbed. When radiation intensity is low, thicker sections may be necessary to obtain sufficient exposure in the requisite length of time. It is difficult to prepare frozen sections as thin as 5 to 10  $\mu$ .

**14.3. Radioautographic Emulsions.** Photographic emulsions available for radioautography may be divided into four general groups according to the application for which they are intended: those used with visible light, x-ray, spectroscopic, and special particle plates. For alpha irradiation the very fine-grained emulsions of the last two groups are most useful, especially the "alpha-particle" film. For beta radiation, however, these plates are not sensitive enough. X-ray or dental film is very sensitive to beta particles, but the grains are large and irregular and hence definition is poor. The somewhat finer grained lantern-slide plates are a suitable compromise. In general, with a small grain size, the probability of a relatively slow and heavy alpha particle hitting and imposing a latent image on a grain is greater than that of a faster beta particle. The probability of a hit with a beta particle, and therefore the sensitivity, increases with increasing grain size, but the resolution diminishes.

As charged particles pass through the emulsion they will be scattered by the atoms of the emulsion. Hence with thicker emulsions, the scattering will be greater and the resolution consequently diminished. The backing or film support also contributes to this effect especially in the case of beta particles of high energy. The usual thickness of x-ray film and lantern-slide plates is approximately 100  $\mu$ , and alpha-particle plates have an emulsion thickness of about 40  $\mu$ . A thinner film may be obtained by using liquid emulsion or by the use of plates recently developed by the Eastman Kodak Company which have emulsion thicknesses of 5 to 10  $\mu$ , which are available under the designation NTP. The use of liquid emulsion or stripping film eliminates scattering due to film backing. On the other hand, if the number of grains is increased by increasing the emulsion thickness, the probability of a beta particle imposing a latent image increases. Therefore when sensitivity and range of response are the desiderata, a thick emulsion should be used.

Exposure and development are the remaining conditions to be considered in the preparation of a radioautograph. The time of exposure will depend to a great extent on the amount of radiation present in the samples to be analyzed. This may be determined prior to preparation of the radioautograph by determining the activity of one or of a few sections by a suitable counter. In general, a satisfactory beta autograph may be obtained with about  $10^7$  particles per square centimeter, whereas  $2 \times 10^6$  alpha particles will be sufficient. However, less total radiation is often sufficient if local concentrations are high. This is also true in the case of alpha autographs made for the study of tracks rather than film blackening. When possible, a number of plates should be exposed and developed at various intervals to enable selection of the most suitable exposure time. In the case of beta autographs made on lantern-slide emulsions, a prior run with the more

sensitive and faster x-ray film will indicate the proper exposure time. Where other factors permit, exposure time should be less than 2 to 3 half-lives of the radioisotope since the latent image fades with time, a phenomenon that may be slowed by storing the setup at low temperature. Although a longer exposure time increases the range of response, the resolution is thereby decreased and the possibility of background fog from stray radiation is increased.

Background fog is a serious problem in cases of low-intensity radiation and in attempting to estimate the quantity of radioactive material by numbers of developed grains. It may be controlled to some extent by careful selection of emulsion and, in some cases, by overexposure and underdevelopment. A permanganate wash during development also diminishes background, but care must be taken since it also decreases image intensity.

In summary, the conditions affecting the three desirable factors are as follows: (1) Sensitivity is increased by use of a thick emulsion of large grain size, by prolonged exposure, and by reduction of preparation time to a minimum. (2) Resolution is improved by careful selection of the proper emulsion for a particular type of irradiation, by use of a thin emulsion with small grain size, by keeping exposure time to a minimum, by decreasing background fog by overexposure and underdevelopment, and through the use of proper chemicals. (3) Adequate range of response is dependent on use of thick emulsions, complete development, and comparison of radioautographs made with various exposure times. Other conditions that are of general value in obtaining good radioautographs include the following: the distance between the sample to be studied and the emulsion must be reduced to a minimum; the radioactive material must not be appreciably soluble in any of the reagents used prior to exposure of emulsion to section; the stain used for the tissue must yield good differentiation, *i.e.*, if applied after development it should not obscure or fade the image, and if applied before exposure it should not be washed out by any of the reagents used for development.

References 1, 2, and 3 are excellent reviews with extensive bibliographies, but a number of the more recent references dealing with the general subject of radioautography will be given here. The physical principles are discussed in papers 4 and 5. The technique of making radioautographs with section and emulsion supported individually and realigned after separate staining and developing is described in 4 and 6. Articles 7 and 8 deal with preparation of liquid emulsions and methods of spreading these on sections. The method of floating section onto emulsion is detailed in references 9 and 10.

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## CHAPTER 15

### THEORY OF TRACER METHODS

**15.1. Introduction.** The multitude and diversity of applications to which tracers have been put make a detailed description of tracer methods impracticable in the present volume. The discussion here will be confined, therefore, to some of the principles of tracer methods which have found the most frequent use. On the other hand, an exhaustive treatment of the possible mathematical descriptions of biological, chemical, and physical systems in which tracers could be utilized would in any case perhaps not be warranted since it would present nothing essentially new. A tagging agent is assumed to be chemically and physically indistinguishable to the system and hence will follow, in most instances, processes already described in great detail in standard references on physics, chemistry, and especially thermodynamics.

Labeling agents are most commonly radioactive isotopes and rare stable isotopes, but they include, as well, substances such as dyes. It is required only that the tracer be chemically and physically exactly equivalent to the substance it represents or displaces and that it in no appreciable way affect the system differently from its normal counterpart. Although distinguishable to the observer, the labeled substance cannot be discriminated by the system so that tagged and untagged molecules enter each process with equal probability. In certain instances this condition is not altogether satisfied. Thus, in dynamic systems in which  $H^2$  or  $H^3$  is introduced as a tracer for hydrogen, or  $C^{13}$  or  $C^{14}$  for  $C^{12}$ , it is known that some discrimination occurs because of the large mass differences in the isotopes. Particularly in tracer experiments with hydrogen isotopes it is necessary to give special attention to possible differential diffusion and reaction rates. Such isotope effects are less pronounced with carbon and are probably entirely negligible for isotopic tracers of greater mass. Very often the tracer must be present in minute quantities to satisfy the condition of indistinguishability. This is especially important with radioactive tracers. A high radiation density produced by excessive amounts of a radioactive tracer may have a profound effect on the character and dynamics of the system into which it is introduced. In order to guard against the influence of excessive radiation it is sometimes desirable to estimate, if possible, the maximum doses to be expected in various parts of the system for a given quantity of radioactive tracer. A third possibility of disturbing the system is encountered when



the substance to be traced occurs normally in minute amounts. It is sometimes found, particularly in biological systems, that the addition of labeled substance considerably in excess of the normal amount present leads to processes that are not characteristic of the normal system. The possibility that a labeling agent may not behave strictly as a tracer should always be kept in mind in any novel application of tracers; caution should be exercised in interpreting experimental results until it has been ascertained that the tracer itself does not appreciably influence the system.

The choice between stable and radioactive isotopic tracers is usually not difficult to make; if a radioactive isotope of the element to be traced is available, it should be used. Radioactive isotopes are, from the technical point of view, by far the easier to detect. A great variety of radiation measuring instruments are available at reasonable cost. Samples are relatively easily prepared, and measurements of specific activity are rapidly made. On the other hand, with the exception of deuterium, stable isotopic tracers can be measured only with the mass spectrometer. The preparation of samples is tedious and in many instances extremely difficult. Both the sample preparation and actual measurement are, in addition, time-consuming and often subject to many uncertainties.

Perhaps equally important is the relative sensitivity of stable and radioactive tracers. The dilution of a stable isotopic tracer in a system is limited by the normal abundance of the isotope and inherent accuracy of the measurements. Mass spectrometers usually have an accuracy of about 1 per cent, or somewhat better under favorable conditions. If, for example, the normal isotopic abundance is 1 per cent, as in the case of  $C^{13}$ , the greatest possible dilution factor is 10,000 and in practice is likely to be more nearly 1,000. For radioactive isotopes, however, dilution factors greater than  $10^6$  are not uncommon in routine practice. Quantities as large as 1 millicurie can sometimes be used in tracer experiments, and quantities as small as  $10^{-4}$  microcuries are always measurable. There remain finally the absolute quantities of the two types of tracers which must be used. Whereas the mass of a tracer quantity of radioactive isotope is negligible by ordinary standards, the mass of a stable isotope necessary to label a substance is a considerable fraction of the total amount of the element present.

In several notable instances it is necessary to resort to stable isotopes. In biological investigations the most important of these are, of course, oxygen and nitrogen. There are no useful radioactive isotopes of these elements, and the stable forms  $N^{15}$  and  $O^{18}$  must be used.

In general, tracer applications are as extensive as the processes and systems they are used to investigate. In the broadest sense, their applications can, however, be divided into three categories: (1) quantitative determinations of substances in complex systems, primarily by the method of isotope dilu-

tion; (2) identification of substances in various parts of complex systems and of the routes that the tagged substances undertake and the processes they undergo; and (3) determination of the rates of transfer and the quantities of labeled substances in various processes and phases of a system. A partial description of some of the methods that can be employed in these applications is given in the following sections.

Many species of radioactive and stable isotopes are available to qualified persons and institutions from the United States Atomic Energy Commission for research, medical, and industrial use. Some of the isotopes are available in a variety of chemical and physical forms and at a moderate price based on the cost of manufacturing and handling. All inquiries relevant to the procurement of isotopes should be addressed to Isotopes Division, United States Atomic Energy Commission, P. O. Box E, Oak Ridge, Tenn.

The available radioisotopes are listed in the *Radioisotopes Catalog and Price List* which may be obtained from the Isotopes Division. Application for any of the listed radioisotopes is made by filing with the Isotopes Division, three copies of *Application for Radioisotope Procurement, A. E. C. form 313*. When the proposed use of the isotope is approved, a purchase order may be sent on receipt of *Authorization for Radioisotope Procurement, A. E. C. form 374*. Form 313 may also be used for procurement of certain organic compounds labeled with the radioisotopes  $C^{14}$ ,  $P^{32}$ ,  $S^{35}$ ,  $I^{131}$ , and  $Au^{195}$ . Inquiries concerning specifications other than those listed in the catalog, and pile-irradiation of substances supplied by the applicant should be addressed to the Isotopes Division prior to preparation of the final form.

Stable isotopes that are available from the United States Atomic Energy Commission are listed in the catalog *Stable Isotopes* or in circular E-13. These isotopes are obtained by preparing the set of forms *Stable Isotope Request form 100*. The isotopes D (and  $D_2O$ ),  $B^{10}$ , and  $O^{18}$  are available for direct purchase, whereas all other stable species, which are separated electromagnetically, can be obtained only on loan.

All isotopes now being sold are also being distributed without charge to qualified physicians and research workers in the United States for use in cancer research, diagnosis, and treatment. The physician who is planning to use the isotopes must have had clinical experience with radiations and must be associated with an institution that is properly equipped to handle radioisotopes. Further details on the conditions under which free isotopes are allocated are contained in circulars E-35 and D-4 issued by the Isotopes Division.

**15.2. Isotope Dilution.** The method of isotope dilution provides a powerful but simple means of quantitative analysis in many applications where all other methods fail or may be extremely difficult. Although it has its own limitations and in many instances is not so convenient as standard analytical

procedures, its particular usefulness is to be found in problems where the total quantity of a substance must be ascertained without at the same time disturbing the system in which it occurs, such as determinations of total body water, or of certain amino acids in humans *in vivo*, or in problems where quantitative recovery together with a high order of purity is otherwise impossible.

In principle the method consists in adding to the system containing the substance to be analyzed a small quantity of the same substance that has been tagged with a radioactive or rare stable isotope. After complete mixing has taken place, a sample of the substance is isolated with the requisite purity and analyzed for its content of the tracer isotope. The dilution of the isotope is then directly related to the total quantity of the substance with which mixing can occur. In practice the major difficulty sometimes lies in synthesizing with a labeling agent the compound to be used. Furthermore, for measurements *in vivo* it is necessary that the substance is neither eliminated nor produced in appreciable quantity during the course of the experiment; *i.e.*, the system must be in static or quasi-static equilibrium during the experiment. In simple solutions and mixtures this difficulty usually does not exist, but in metabolizing systems some caution must be exercised.

When the labeling agent is a radioactive isotope, computation of the total initial quantity of substance in the system from the observed dilution is simple. Denoting the specific activity of the material introduced by  $x_1$  and that of the sample afterward taken from the system by  $x_2$ , the total quantity of diluent [1,2] is

$$Q = q \left( \frac{x_1}{x_2} - 1 \right) \quad \text{gm or cc}$$

where  $q$  is the quantity of labeled material added.

The situation with regard to stable isotopic tracers is somewhat more involved so far as measuring the isotopic concentrations is concerned. Both diluent and labeled material contain the two isotopic species; the former contains both isotopes in their normal abundance, and in the latter the rare isotope is enriched by an arbitrary but accurately known amount. For this reason it is more convenient to express isotopic concentrations in atom per cent excess (per cent in excess of normal abundance) rather than atom per cent. In most applications of the dilution method there is only a single diluent and carrier (labeled substance), and only two isotopes to consider. The total quantity of diluent [1,2] is then

$$Q = q \frac{M_1}{M_2} \left( \frac{C_1}{C_2} - 1 \right) \quad \text{gm or cc}$$

where  $q$  = grams of labeled substance added

$M_1$  = molecular weight of added substance with enriched rarer isotopes

$M_2$  = molecular weight of diluent with normal isotopic composition

$C_1, C_2$  = atom per cent excess of rare isotope in carrier before adding and after recovery, respectively

If the molecular weight is large and the tracer isotope occupies only a few atomic positions in the molecule, then  $M_1/M_2 \approx 1$ .

**15.3. Tracer Problems Involving First-order Reactions.** Many of the tracer problems encountered at the present time, particularly in biological investigations, are concerned not only with identification of constituents and metabolic routes in systems of widely varying degrees of complexity but also with kinetic properties of systems. Although measurements of the amounts of a labeling agent in various parts of a system serve sufficiently well to identify constituents and chemical or physical processes, some kind of mathematical formulation based on the experimental data is required to describe adequately the rates of processes. The precise method of formulating the appropriate mathematical expression may not, of course, always be immediately obvious since, presumably, it may include any of the processes already known to chemical kinetics.

When the processes under consideration can be shown to be first-order reactions their mathematical description becomes particularly simple and, needless to say, extremely useful. Reactions of this type proceed at rates proportional to the amount of substance present. They may be described, therefore, by linear first-order differential equations of the type

$$\frac{dx}{dt} = k_1x - k_2f$$

where  $x$  is the amount of substance present at time  $t$ ,  $k_1$ , and  $k_2$  are constants, and  $f$  is some arbitrary function (arbitrary in a mathematical sense). The first term on the right is the amount added per unit time, and the second term represents the amount disappearing per unit time. A considerable simplification is usually introduced in practice in that the system with its manifold processes is in dynamic equilibrium during the time it is investigated. The differential  $dx/dt = 0$ , and the differential equation then becomes an algebraic equation. Within the system substances may be produced and utilized, transferred from one region or tissue to others, or undergo changes in chemical and physical form; but all such processes take place at constant rates. The addition of a small quantity of material, homologous with a normal metabolite already present and labeled with a convenient tracer, will not appreciably disturb the system and presumably will undergo the same processes without discrimination. The tagged molecules themselves, however, will not be in equilibrium, and since they are

distinguishable to the observer, their change in concentration is described by the differential equation above. It is for this reason that tracers provide a powerful tool for investigating rates of processes in dynamic systems. The observed change in concentration of tagged molecules, or in some cases, the labeling agent itself, is in each process directly related to the constant rate of reaction for the untagged homologue, or more exactly, for the total tagged plus untagged material.

A great variety of first-order physical and chemical processes may be described by identical mathematical expressions in which only the units differ. These processes may include such apparently divergent phenomena as the transfer of a labeled substance from one region or tissue to another, chemical reactions in which a substance  $A$  is degraded or synthesized into a series of substances  $A \rightarrow B \rightarrow C \rightarrow \dots$ , radioactive decay, absorption of gamma rays, and combinations of these and other processes. For the sake of generality, therefore, a labeling agent introduced into a system in one form or another will be considered to be present in certain "phases" with the understanding that a phase may refer to a particular chemical or physical form, an organ or certain tissue, or a specified volume with or without well-defined geometrical form. The precise meaning and units of phase will, consequently, depend upon the type of system and process under consideration. Expressed in general terms, tracer techniques provide the means for identifying the phases involved in the processes under consideration, determination of the amounts of substance present in each phase, and the determination of the rates of change of a substance from one phase to another.

For a steady-state system containing only irreversible first-order reactions, the concentration of a labeled substance as a function of time in any one phase of the system can be represented by the polynomial

$$x = a_1 e^{k_1 t} + a_2 e^{k_2 t} + \dots + a_n e^{k_n t}$$

The parameters  $a_i$  and  $k_i$  can be determined only from measurements of the tagged molecules in the phase. The coefficients  $a_i$  may take any positive and negative values or may be zero. They represent the amplitude of the separate terms when extrapolated back to zero time. For phases other than the one in which the labeled substance is initially introduced, the coefficients usually have the property that  $a_1 + a_2 + \dots + a_n = 0$ . The unit of  $a_i$  may be microcuries or microcuries per gram when radioactive isotopes are used, and atom per cent or atom per cent excess for stable isotopes.

The parameters  $k_i$  are negative or zero but not positive, since positive values indicate a concentration increasing without limit. They represent the fractional amounts of labeled substances entering (for  $a_i$  positive) and leaving (for  $a_i$  negative) the phase per unit time.

In practice the experimental data, consisting of periodic measurements of

the tracer concentration, are plotted on semilog paper with a linear time axis. The separate exponential component of the resulting curve may then be subtracted out as straight lines. From the analyzed curve the coefficients  $a_j$  are obtained by extrapolating each component back to zero time. The parameters  $k_j$  are also obtained from the component curves by their relation to the half-times,  $k_j = 0.693/T_j$ . For most purposes the half-times  $T_j$  are determined with sufficient accuracy by inspection. However, unless the

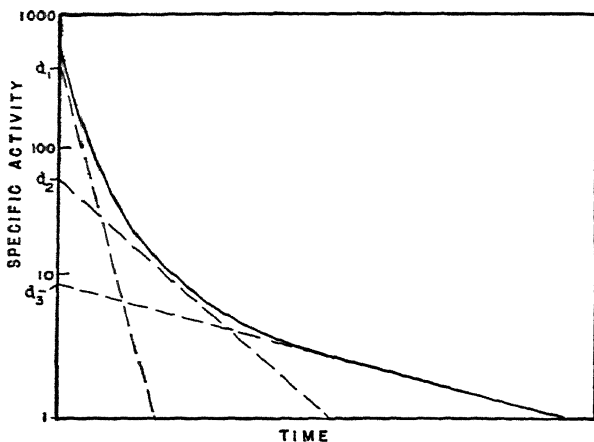


FIG. 110. Reduction of elimination curves of the type  $x = a_1e^{-k_1t} + a_2e^{-k_2t} + \dots$  into separate components. Three components are shown in the diagram together with the curve from which they are reduced. The longest component is taken out first by approximating the straight line to which the curve is asymptotic. The remaining components are obtained by successive subtractions.

parameters  $k_j$  differ in magnitude by a factor of two or more, it is often difficult to evaluate them, and also  $a_j$ , with accuracy or even to assign uniquely the number of components that appear to be present. This becomes especially difficult when the number of terms exceeds three or four. In any event, whether one or several components are present the physical and physiological significance of the constants may not always be obvious until other phases of the system are also investigated.

A number of simple processes frequently encountered in tracer studies of biological systems are described in greater detail below. It must be kept in mind, however, that the expressions are valid only for systems possessing the following physical characteristics: (1) only first-order reactions occur; (2) the system cannot distinguish between labeled and unlabeled substance of the same chemical and physical form; (3) the system, except for the tagged molecules, remains in a steady state; (4) the mixing time of tagged molecules

with untagged molecules of the same substance in any phase is short compared to the time in which the concentration changes appreciably after equilibrium.

*a. Simple Elimination from One Phase.* The total quantity  $M$  of substance in the phase is assumed to remain constant; the rates, in either grams or cubic centimeters per unit time, of appearance and disappearance are equal and constant. If labeled material of the same form is added to make the specific activity in the phase at  $t = 0$  equal to  $X$  microcurie per gm, the specific activity subsequently decreases as

$$x = Xe^{-kt}$$

The parameter  $k$ , obtained from the plotted measurements, is referred to as the turnover rate for the substance being traced. Physically, it is the fraction  $r/M$  of the total amount of the substance (labeled plus unlabeled) replaced per unit time. Its reciprocal  $1/k = M/r$ , called the turnover time, is the time required for the replacement of an amount equal to  $M$ . The quantity  $r$  is then the actual rate, in grams or cubic centimeters per unit time, at which the substance (tagged plus untagged) enters and leaves the phase. The turnover time  $1/k$  is readily determined by inspection from the experimental data plotted on semilog paper since  $1/k = T/0.693$ , where  $T$  is the half-time for disappearance of the tagged molecules. Thus far only the ratio  $r/M$  has been found. The actual value of  $M$ , and hence of  $r$ , can however be determined, as described in Sec. 15.2, by the dilution of the administered active material (at  $t = 0$ ).

It is apparent that more than one route may be taken by the substance when it leaves the phase, for the turnover rate may be written also as the sum of rates of transfer to several different phases; e.g.,

$$\frac{r}{M} = \frac{(r_1 + r_2 + r_3 + \cdots)}{M}$$

As measured in the one phase, the dilution with time still follows the single exponential expression above since only the sum of the rates is apparent. The separate rates can be determined only if the labeled substance can be traced to the phases that follow.

*b. Labeled Substance Accumulated in One Phase.* Accumulation of substances normally found in a system usually does not occur over extended periods when the system is in a steady state. The important exceptions to this are substances eliminated from the system, usually in expired gases, feces, and urine. In practice excreta are either accumulated over a suitable length of time and the total content of excreted tracer is measured, or the excreta may be sampled periodically for the concentration of the tracer. It is usually not reasonable in such measurements to speak of specific activity,

but rather to consider the actual amount  $u$  of the eliminated (or accumulated) tracer. The rate of elimination (or of accumulation) can sometimes be represented by

$$\frac{\Delta u}{\Delta t} = -a_1 e^{-k_1 t} - \dots + b_1 e^{-h_1 t} + \dots \text{ microcuries/day}$$

The total amount eliminated during the interval of time 0 to  $t$  is then

$$u = -k_1 a_1 (1 - e^{-k_1 t}) - \dots + b_1 h_1 (1 - e^{-h_1 t}) + \dots \text{ microcuries}$$

A second example of accumulation sometimes occurs when an essentially

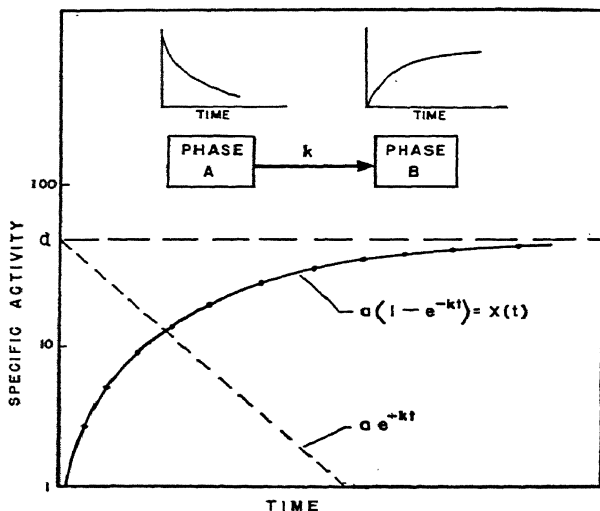


FIG. 111. Simple uptake curve. The uptake curve  $x(t)$  is subtracted from the constant  $a$  to obtain  $e^{-kt}$  from which the parameter  $k$  can be determined by inspection. This method presumes only one component to be present and that measurements are continued long enough to enable a reasonable value of  $a$  to be estimated.

foreign substance is introduced into the system. If it is introduced into phase  $A$  from which it is transferred to phase  $B$  by a first-order reaction, the amount accumulated and fixed in  $B$  is

$$u_b = u_a(1 - e^{-kt}) \quad \text{microcuries}$$

while the amount remaining in  $A$  is  $u_a = u_0 e^{-kt}$ . The specific activity of the substance does not change; the substance is not normal to the system, and the ratio of tagged to untagged molecules remains constant in any process.

*c. Constant Uptake Rate and Exponential Elimination.* When the rate of uptake of labeled molecules is maintained at a constant value of  $p$  micro-



curies per sec. but the rate of disappearance from the phase is proportional to the concentration, the specific activity, assuming it is zero initially, is

$$x = \frac{p}{kM} (1 - e^{-kt}) \quad \text{microcuries/gm}$$

As before,  $k$  is the turnover rate or  $k = r/M$ , where  $M$  is the total constant quantity of substance in the phase and  $r$  is the constant rate of disappearance. After a long time compared to  $1/k$ , the specific activity approaches the constant value of  $p/kM$ .

*d. Transfer between Two Phases.* One of the most common rate problems investigated with tracer techniques is the transfer of a labeled substance or of the labeling agent itself from phase  $A$ , into which it is introduced, to one or more phases  $B, C, D, \dots$  where the activity is measured. It is assumed as before that the system is in a steady state, the amount  $M$  of the substance (labeled plus unlabeled) in each phase is constant, and the rates  $r$ , of appearance and disappearance of the untagged substances are constant.

Consider  $A$  to be the precursor of several phases  $B, C, D, \dots$  which receive material directly from  $A$ . If the specific activity in phase  $A$  is raised initially to  $X$  microcuries per gm of labeled substance, its subsequent specific activity is  $x_1 = Xe^{-k_1t}$ , where  $k_1 = r_1/M_1$  is the turnover rate,  $M_1$  is the total quantity, and  $r_1$  is the rate of disappearance from  $A$ . The specific activity in any one of the phases  $B, C$ , or  $D$ , for which the turnover rate is

$$k_2 = \frac{r_2}{M_2},$$

is initially zero but after a time  $t$  it is

$$x_2 = \frac{k_2 X M_1}{(k_2 - k_1) M_2} (e^{-k_1 t} - e^{-k_2 t}) \quad \text{microcuries/gm}$$

The activity in this phase increases from zero to a maximum in the time

$$t_{\max} = \frac{1}{k_1 - k_2} \log \frac{k_1}{k_2}$$

Afterward the concentration of the tagged molecules decreases at a rate equal

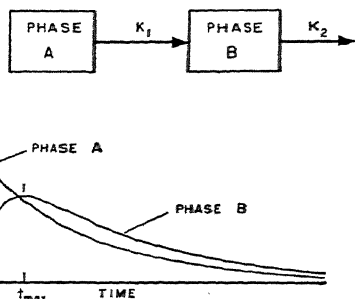


FIG. 112. Specific activities in separate phases  $A$  and  $B$  in a simple two-phase system as a function of time.

to the smaller of  $k_1$  and  $k_2$ . The expression above for  $x_2$  is equally valid when  $A$  is the precursor of only  $B$  and all the material that leaves  $A$  reappears, usually as a different chemical form, in  $B$ .

It is apparent from inspection of the formula above that when  $k_1 = k_2$  it is no longer valid. In this special case it takes the indeterminate form  $0/0$  and must, therefore, be used in its limiting form when  $k_1 \rightarrow k_2$  which is

$$x_2 = \frac{k_1 X M_1}{M_2} t e^{-k t} \quad \text{microcuries/gm}$$

*e. Series of Three-phase Changes.* However complex a system may be, it is often possible to investigate a specific phase of it by introducing labeled

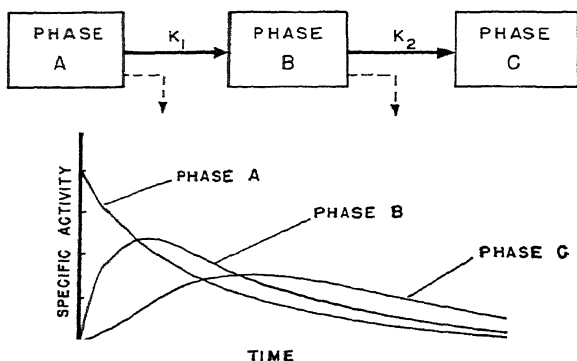
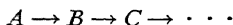


FIG. 113. Uptake and elimination of tagged material in three phases. The curves indicate the time variation of specific activity in each phase when the labeled material, introduced at phase  $A$ , passes directly through all three phases. It is possible in certain instances for the labeled material to leave also by other routes as indicated by the broken lines. The areas under the curves will then be somewhat smaller, each by a different factor, than if all the material were to pass from  $A$  to  $C$ .

material directly into the phase or into the immediately preceding phase. If it is necessary to go back two or more phases, the reactions, particularly in biological systems, tend to become exceedingly complex. When, however, a substance in phase  $A$  passes successively through a series of phases



and only  $A$  can be tagged, the concentration of the tagged molecules in  $C$  can be described by an expression similar to those above. After introducing into  $A$  an amount of labeled substance to raise the specific activity to  $X$  microcuries per gm, the material, possibly in different chemical forms, passes from  $A$  to  $B$ ,  $B$  to  $C$ , and from  $C$  it is eliminated or otherwise metabolized.

The activity in  $C$  at any time  $t$  is then

$$x_3 = \frac{k_1 k_2 X}{(k_2 - k_1)(k_3 - k_1)(k_3 - k_2)} \frac{M_1}{M_3} \left[ (k_3 - k_2)e^{-k_1 t} - (k_3 - k_1)e^{-k_2 t} - (k_1 - k_2)e^{-k_3 t} \right] \text{ microcuries/gm}$$

where  $k_1, k_2, k_3$  = turnover rates of phases  $A, B$  and  $C$

$M_1, M_3$  = total amounts of traced substances in phases  $A$  and  $C$ , respectively

It is seen that this formula is a form of the general expression consisting of a polynomial with exponential terms. In this case the explicit expressions for the coefficient  $a_i$  are rather complicated because of their interdependence on the turnover rates  $k_1, k_2$ , and  $k_3$ .

A classic example of the usefulness of the formulas outlined above for describing one-, two-, and three-phase systems is provided by the experiments of Zilversmit and his associates [6,7]. Radioactive phosphorus ( $P^{32}$ ) was used in these experiments to determine the turnover rate of phospholipids in the plasma of dogs. Measurements of the specific activity of the phospholipids and of their immediate precursor gave the necessary curves representing the uptake and disappearance of tagged molecules from which, by analysis similar to that above, the turnover rate of phospholipids could be determined.

**15.4. A More General Theory of Tracer Methods.** While the descriptions of processes given by the simple expressions for first-order reactions are probably the most widely useful, they are by no means adequate for systems in which higher order reactions are involved. When it is found advantageous to describe such processes in terms of mathematical expressions, it may be possible, as in the case of first-order reactions, to formulate the requisite differential equation and to solve for a particular solution in terms of suitable initial and boundary conditions. This procedure, however, is possible only when the nature of the process is already known in some detail. On the other hand, general equations may be found that are valid for large classes of phenomena. Thus, in first-order reactions the concentration of a tracer

as a function of time is given by the general formula  $x = \sum_{i=0}^n a_i e^{k_i t}$ . A more

general equation has been pointed out by Branson [3] who showed its usefulness and great power when applied to tracer investigations of metabolizing systems. In view of the importance of tracer applications to biological systems, the equation and a few of its uses described by Branson are given below.

A generalized metabolizing system may be regarded as a single complex phase in the sense described in the last section. The phase may be a physical

or chemical state, or it may be a specific tissue or a region, with or without well-defined boundaries, in which the substance under consideration is consumed, produced, transferred, modified, or stored. Despite a great variety of processes that may conceivably occur, these systems can be described by an integral equation already familiar to mathematicians [4,5].

In order to clarify the meaning of the equation, consider a single phase in which the total amount of metabolite present initially ( $t = 0$ ) is  $M_0$ . Since it is being metabolized, there remains of this original amount after a time  $t$  the quantity  $M_0 F(t)$ , where  $F(t)$  is some metabolizing function appropriate to the system and a function only of time. Simultaneously, additional molecules of metabolite are accumulated at the rate  $R(t)$ , and in any interval of time  $\theta$  to  $\theta + d\theta$  an amount  $R(\theta) d\theta$  is added. The accumulating metabolite also undergoes metabolism, and of the amount  $R d\theta$  added at time  $\theta$ , there remains at time  $t$  the amount  $R(\theta)F(t - \theta) d\theta$ . The total quantity of metabolite in the phase at time  $t$  is then the sum of  $M_0 F$  plus what remains of the amount added during 0 to  $t$ , giving the general integral equation

$$M(t) = M_0 F(t) + \int_0^t R(\theta) F(t - \theta) d\theta$$

The method of solving the equation depends on which of the functions  $M(t)$ ,  $M_0$ ,  $F(t)$ , and  $R(t)$  are known or can be determined empirically. When  $M_0$ ,  $R(t)$ , and  $F(t)$  are known,  $M(t)$  can, of course, be obtained by direct or numerical integration. If  $M(t)$ ,  $M_0$ , and  $F(t)$  can be found, the unknown function  $R(t)$  appears only in the integral and the equation becomes a Volterra integral equation of the first kind in  $R$ . Finally, when all the functions but  $F(t)$  are known, it becomes a Volterra integral equation of the second kind in  $F(t)$ .

In certain cases the metabolizing system will permit simplifications to be made in the equation above. Thus, if the system is in dynamic equilibrium, the total amount  $M(t)$  of metabolite present remains constant, or  $M(t) = M_0$ , and the integral equation then becomes

$$M_0(1 - F(t)) = \int_0^t R(\theta) F(t - \theta) d\theta$$

In some instances, also, the rate  $R$  is known to be constant or, if not, it may sometimes be held constant during the course of the experiment. When simplifications such as these are possible, the work of determining the unknown functions and solving the integral equation is greatly facilitated.

Empirical determination of the functions depends upon which quantities are accessible to measurement without seriously disturbing the system. For this purpose tracer methods are perhaps the most powerful. While the labeled homologue of the normal metabolite will also be described by the integral equations above, the functions  $M(t)$  and  $M_0$  will not, in general, be

the same for the tagged and untagged fractions. However, if it can be assumed that the system does not discriminate between tagged and untagged molecules, the functions  $R$  and  $F$  will be the same for both; thus, measurement of the change in concentration of tagged molecules in the phase enables a choice of functions  $R$  and  $F$  to be made which will, therefore, describe the kinematic behavior of the normal metabolite. The procedure to be followed may be illustrated by two applications described by Branson. The first method assumes that the metabolite can be labeled and introduced as a single dose at  $t = 0$ . The equation for the tagged metabolite then reduces to the simple form  $M^*(t) = M_o^*F(t)$ , where  $*$  denotes the labeled metabolite.  $F(t)$  is determined directly from measurements of  $M_o^*(t)$  and  $M^*$ , and when substituted in the integral equation

$$M(t) = M_o F + \int_0^t R F d\theta$$

$M(t)$  can be found by integration if  $M_o$  and  $R$  are known, or the equation may be solved for  $R$  if  $M(t)$  and  $M_o$  can be determined empirically. Usually,  $M(t) = M_o = \text{constant}$  which can be determined by the method of dilution.

The second case is a considerably more complex problem that arises when two or more phases must be considered, as when the precursor  $A$  of the metabolite  $B$  is labeled. If a single dose of labeled  $A$  is added, the steady state system  $A \rightarrow B \rightarrow$  is described by the equations

$$\begin{aligned} A^*(t) &= A_o^* F_1 \\ A(1 - F_1) &= \int_0^t R_1 F_1 d\theta \\ B^*(t) &= \int_0^t R_2 F_2 d\theta \\ B(1 - F_2) &= \int_0^t R_2 F_2 d\theta = B^*(t) \end{aligned}$$

An additional relation  $B^*(t) = B_o^* F$  can be used if the metabolite can be tagged and introduced into phase  $B$  in the same or a similar system but in a separate experiment when  $B^*$  from the precursor is not present. Alternatively a second labeling agent, e.g., a stable isotope, or a different species of radioactive isotope distinguishable from the first, may be used to determine the relation  $\bar{B}(t) = \bar{B}_o F_2$ , where the bar indicates the metabolite tagged with the second isotope. Although technically the latter procedure may be, and usually is, considerably more difficult, it provides a distinct advantage in that all the quantities required may be measured simultaneously and in the same system.

The general procedure to be followed in applying the integral equation to complex systems of several interrelated phases is found as an extension of the example above. The metabolite  $M_i$  in any phase  $i$  is described by the

general integral equation given above, with  $n$  equations for the  $n$  related phases. To these are added the equations for the labeled substances. The form of these equations can be simplified to some extent by proper choice of the experimental conditions. Thus, if the substance  $M_i$  is labeled with an isotope, designated by the subscript  $j$ , and introduced into phase  $i$ , relations of the form

$$M_{ij}^*(t) = M_{ij}^* F_i$$

are obtained, where  $F_i$  is the metabolizing function of the phase and is not influenced by the particular isotopic label. In addition, if the precursor of  $M_i$  is labeled with the isotopic tracer  $k$ , a set of relations are obtained in the form

$$M_{ik}^*(t) = \int_0^t R_i F_i d\theta$$

In each such phase  $(M_o)_{ik}^*$  is zero since initially no labeled metabolite is present and none appears until it is provided by the precursor through normal processes.

For most of the elements of biological interest, there are now available several species of isotopes, e.g.,  $H^2$  and  $H^3$ ,  $C^{13}$  and  $C^{14}$ , and the several useful isotopes of iron. While the technical difficulties often increase rapidly with the multiplicity of tracers used in a single experiment, the more extended field of applications open to multiple-tracer techniques in complex systems should not be overlooked. The technique is still relatively undeveloped, but already many instances are found where systems are accessible to detailed investigation only through such techniques and, perhaps as well, through more advanced mathematical treatment such as that outlined above.

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## CHAPTER 16

### INTERNAL DOSIMETRY

**16.1. Physical Principles of Dosimetry.** The essential purpose of dosimetry is the quantitative evaluation of some effect produced in tissue by a given quantity and type of radiation. The radiations with which internal dosimetry is chiefly concerned are those emitted by radioactive materials distributed in or near tissue. The principles and methods outlined in the following sections, however, apply equally well to other kinds and sources of radiation including x-rays and neutrons, protons, deuterons, and alpha particles obtained from high-energy accelerators.

The first requirement of dosimetry is an index on which a unit of dose can be established. In the most general terms the index may be a biological indicator, some physical or chemical effect produced by the radiation, or some property of the radiation itself. The ultimate information desired is, in nearly all cases, the biological or clinical effects produced by a given radiation or source of radiation. As yet, however, no biological indicator has proved satisfactory as a unit of dose. Factors such as mean lethal dose, chromosomal changes, and mitotic activity may be taken as biological indicators but are useless as units of dose. They do not lend themselves to convenient measurement and not at all to calculation. Different tissues and organs as well as different species of animals vary widely in their resistance to radiation so far as observable biological or clinical results manifest themselves. Moreover, for any one tissue exposed to a given energy flux of radiation (mev or ergs per square centimeter per second), observable biological effects are strongly dependent on both the type and the energy of the radiation. For these reasons an index based on any one biological effect would in practice be difficult to correlate with other effects and with similar processes in different tissues. This applies equally well to chemical effects induced by radiation since they are subject to the same shortcomings. There remain, consequently, only physical properties of either the radiation itself or its interaction with tissue.

From a physical point of view the ideal unit of dose is either the energy absorbed from the radiation per unit mass of tissue or the ionization formed per unit mass. Both these quantities can be measured with reasonable accuracy and may be expressed in absolute units. Since units of energy

absorbed and of ionization may be defined so that they are more or less independent of the type of radiation and absorbing medium, dose expressed in such physical units may be accurately reproduced in different tissue and can be correlated with any biological or clinical effects that occur.

Units of energy absorbed per unit mass or volume of medium have, at the present time, found universal although informal acceptance for expressing dose delivered by corpuscular radiations. The important features of such units are their evaluation in terms of fundamental physical units, the accuracy and convenience with which they are measured, at least in principle, and the fact that they are amenable to calculation when the source of radiation is given. On the other hand, the ionization produced per unit mass of tissue possesses essentially the same qualifications as energy for a unit of dose in addition to the important physical distinction of being the immediate and only directly observable product of radiation interaction with tissue. Ionization as such is in effect the only measurable precursor of the biological, clinical, and chemical effects that occur. From the standpoint of dose measurement, some advantage is to be found in units based on the number of ion pairs formed per unit mass since the dose is then given in terms of the quantity directly measured with the ionization chamber.

The units of energy absorbed which have found the most widespread use include the roentgen-equivalent-physical (U. S.), the gram-roentgen (British), and the energy-unit (British). The only unit of ionization is the J, proposed by the British Committee for Radiological Units. The roentgen, which applies only to x- and gamma radiation, is actually a partial description of the radiation at any specified point (not necessarily in an absorbing medium), but it is regarded by some as essentially a unit of energy absorbed in air, *i.e.*, 83 ergs per gm of air—a meaning that is not strictly correct.

Whichever unit of dose is employed, the physical quantity actually measured is, in all instances, the ionization produced per unit volume per unit time. In the case of dose expressed in roentgens, the ionization measured must, by the definition of the roentgen, be that produced in air at the point under consideration. In order to make use of conveniently small ionization chambers and yet measure true air ionization, it is necessary to use "air-wall" chambers, *i.e.*, chambers with solid wall material consisting of airlike atomic composition, in determining dose in roentgens. Dose expressed in the various energy units requires conversion of the detected ionization to ergs or mev of energy absorbed. This can be done only when the average energy  $W$  required to form one ion pair in the medium is known. For ions produced in air by electrons,  $W = 32.5$  ev. In the absence of conclusive experimental data on tissue, this value is sometimes adapted to tissue dose assuming air and tissue to be nearly the same in atomic stopping power for electrons.



Tissue dose obtained by calculation is most conveniently stated in terms of one of the units of energy absorbed (ergs, mev, rep, or gram roentgens) since calculations are based directly on the energy released by disintegration of a radioactive isotope or on the energy flux (ergs or mev per square centimeter per second) of beams of neutrons, protons, or other radiations. Conversion of energy absorbed to ionization units, and also to roentgens in the case of gamma radiation, again requires that the value of  $W$  be known for the medium.

A distinction, sometimes overlooked, should be made in the terms dose, dosage rate, total dose, integral dose, and the various combinations of these terms that are frequently used in connection with radioactive isotopes. The term dose refers to roentgens (for gamma rays only), to energy absorbed per gram of tissue, or to ionization produced per gram of tissue at a specified point. These quantities usually vary throughout the volume of an organ or animal in some complicated manner depending upon the source and type of radiation. Since dose is a point function it can often be represented, as in x-ray dosimetry, by a family of isodose surfaces, each surface containing all points in the medium at which the dose has a particular constant value. A statement of dose should, in general, always be qualified by a precise description of the point at which the dose was measured or computed. It must be assumed otherwise that the dose was uniform throughout the organ or entire animal involved. The interval of time in which a specified dose was given is not involved explicitly since all dose units are independent of time.

Time is introduced explicitly only in the term dosage rate, which, depending upon the units chosen, is the roentgens exposure, energy absorbed, or ionization produced per unit time at the point under consideration. For radioactive isotopes the dosage rate usually is not constant but depends in some more or less complicated way on metabolic activity and radioactive decay. It is then convenient to speak of total dose, which is the dose received at a point in the tissue during the whole interval of time the isotope remains in the tissue, or during complete decay of the isotope if the isotope remains fixed in the body.

The terms above refer only to points within the tissue or other medium, and the dose units are roentgens for gamma rays, rep, energy units, or gram roentgens per gram, or J units for any ionizing radiation. Integral dose alone refers to the total energy absorbed or total ionization produced in an entire organ or in a given mass of tissue. The single unit of integral dose is the gram roentgen, and in particular, the roentgen, rep, energy unit and J cannot be used as a unit of integral dose.

Combinations of terms, such as total integral dose and integral dosage rate, are also frequently used.

It was in effect stated above that energy absorbed and ionization produced per unit volume are absolute measures of tissue dose. While this is strictly true within the limits of the definitions of the various dose units, there still remain important uncertainties in the actual measurement of ionization under certain conditions and in both the physical and biological significance of the energy absorbed.

The first difficulty is encountered only in measurements of ionization produced by heavily ionizing particles such as alpha particles and fission fragments. The ionization produced by these particles is extremely intense since their entire energy is spent in ionizing the relatively small volume of tissue enclosing the short path of each particle. The resulting high ion density presents a difficulty in measuring dose with an ionization chamber in that partial recombination tends to take place among ions of opposite charge before they have been separated and swept out of the gas volume of the ionization chamber by the electrostatic collecting field. It is likely that something less than the total ionization produced by multiply-charged particles is actually detected, whereas, for beta particles whose ionization is more dispersed, this is not the case except at very high pressure ( $> 10$  atm).

The local high density of ionization caused by these radiations also leads to more profound biological and clinical effects than are observed for the same energy absorption or ionization produced per cubic centimeter of tissue exposed to beta particles and gamma rays. Greater biological effectiveness of a similar magnitude is also found for protons, deuterons, and for fast and slow neutrons. Indeed, there is reason to believe that for the same dose the biological effectiveness of beta particles varies with energy, although to a lesser extent than the difference between beta particles and protons or alpha particles. It is also well known that even for electromagnetic radiation clinical effects vary with the energy composition of x-ray beam. At the present time these effects can only be lumped under the term relative biological effectiveness (R.B.E.) which expresses the ratio of gamma-ray dose to the dose that is required to produce the same biological effect by the radiation in question. The evaluation of this factor for various radiations is not easy nor can it be extrapolated with certainty from one tissue to another. This is due primarily to the difficulty in evaluating biological effects quantitatively and because of the striking variation in the magnitude of the effects produced in different tissues and animals. The R.B.E. of protons, deuterons, alpha particles, and neutrons has, however, been extensively investigated in certain species of animals, notably the mouse and rat. For these mammals, and possibly also for man, the R.B.E. of protons, deuterons, and neutrons is about 4, while for alpha particles it has a value of about 10, *i.e.*, 0.1 rep of alpha particles produces about the same biological effect as 1 r of x-rays (x-rays not photoelectrically converted). Considerable caution must be

exercised in extrapolating these values to other animals or to specific tissue since, as has been illustrated by extensive experiments with neutrons, values of R.B.E. varying from 0.8 to as much as 30, depending upon the neutron energy and substance irradiated, have been reported by various investigators.

The precise physical significance of the absorbed energy is not as yet entirely clear. The greater part of the energy absorbed from incident radiation is perhaps utilized in the production of ions in the medium, but there is also reason to believe that an appreciable fraction of the absorbed energy is taken up by processes in which no ions are formed, such as excitation of atoms and molecules, decomposition of complex molecules, and the formation of certain substances as in water exposed to x-rays. Although the influence of these processes is not yet well known, they do not directly affect the practical problems of dosimetry since the term dose as used in radiobiology means essentially a measure of the energy absorbed without regard to the manner in which it is absorbed. Ionization, therefore, serves as an empirical measure of energy absorbed.

The empirical determination of dose received by tissue exposed to radiation requires some means for ascertaining the density of ionization within the tissue. This is made possible by measurement of the ionization produced in the gas of a small cavity ionization chamber placed at the point under consideration. A discussion of the principles of the cavity chamber is given in Sec. 12.2, based on the detailed description of the theory and application of such chambers to gamma rays and neutrons reported by Gray [12,18]. In brief, it consists of a small gas-filled cavity with linear dimensions that are small compared to the range of the secondary beta particles in the gas. If the cavity walls are tissuelike in atomic composition or, if differing from tissue, they are made extremely thin and surrounded with tissue, the energy absorbed in the tissue per cubic centimeter of tissue at the point where the cavity is placed is related to the observed gas ionization by the simple relation

$$E = JWS \quad \text{ergs/cc/sec}$$

where  $J$  = number of ion pairs formed in gas per cc per sec

$W$  = average energy absorbed to form one ion pair in gas (about 32.5 ev for electrons in air)

$S$  = stopping power of wall material (tissue) relative to the gas

The factor  $S$  is given by the ratio  $N_t B_t / N_g B_g$ , where  $B_t$  and  $B_g$  are the stopping numbers of tissue and gas, respectively, computed from Bethe's stopping formula, and  $N_t$  and  $N_g$  are the respective numbers of atoms per unit volume of tissue and chamber gas. If the atomic composition of the chamber gas is made similar to that of tissue, then  $B_t = B_g$  and the factor  $S$  is simply the ratio of the number of atoms per unit volume of tissue to the number per unit

volume of gas. For many purposes air is sufficiently similar to tissue to use the simple ratio  $S = N_t/N_{\text{air}}$ ; in more accurate measurements, gas mixtures with approximately the same atomic composition as tissue can be prepared. The average percentages of the principal atomic constituents by weight in lean tissue, for example, are hydrogen, 9.9 per cent; carbon, 13.4 per cent; nitrogen, 4.1 per cent; oxygen, 70.11 per cent; and light minerals, about 1 per cent.

Chambers based on the cavity principle are applicable to all types of radiation and absorbing media. They provide an accurate means for determining dose in terms of absolute quantities, either ionization produced or the energy absorbed expressed in fundamental physical units. When one is measuring the dose in roentgens, the chamber gas and wall material, as pointed out above, cannot be chosen arbitrarily but must consist of air and airline substances.

Calculations of dose delivered to tissue by a uniformly distributed source of charged corpuscular radiation (beta particles, alpha particles, and fission fragments) may be expected under favorable circumstances to yield results in reasonable agreement with the actual dose. Under less favorable conditions and in most instances where x- or gamma radiation is involved, calculations of dose by the methods available at present can be relied upon to give only the order of magnitude of the true dose. The uncertainties accompanying such calculations are due chiefly to the lack of sufficiently detailed information concerning the exact disposition and concentration of the radioactive isotope throughout a tissue and to the difficulty in evaluating that fraction of the energy made available by radioactive decay which is actually absorbed in the tissue under consideration.

The importance of metabolism as a factor in internal dosimetry is obvious. The dose delivered to specific tissues by an administered radioactive isotope depends upon the amount taken up in the tissue and the length of time it remains there. The metabolic paths and rates of turnover are not unique for a given isotope but often depend on the chemical substance in which the isotope is incorporated. These factors are known in some detail for certain organic molecules that can be labeled with active isotopes, and the metabolic fate of a few isotopes such as iodine, phosphorus, and sodium in inorganic salts is reasonably well understood. For a few other substances it is sometimes possible to determine the concentration in a specific tissue either from biopsy or from previous tracer investigations. In most instances the concentration does not remain constant in any one tissue, but, depending upon the complexity of the chemical reactions the administered active substance undergoes, it is regulated by the metabolic rates of uptake and elimination. From the clinical standpoint, estimates of dose are further complicated by the considerable variation among different individuals in the tissue mass of

organs and in the metabolic rates of uptake and elimination. The simplest problem, for example, is encountered when the isotope is rapidly accumulated and fixed in one or more organs, but instances of this are few, and calculations based on this assumption usually do not serve even as a first approximation for most of the problems met with in practice.

The rate at which energy is made available by the decay of a known concentration or total quantity of administered isotope is easily calculated if the decay scheme for the isotope is known. The distinction between available energy and actual decay energy released per disintegration should, however, always be kept in mind. For example, in simple beta decay the available energy per disintegration is not the beta maximum energy given in isotope tables but an average value which is often about one-third of  $E_{\max}$ . The remainder of the decay energy is carried off by neutrinos which do not contribute to the tissue dose. Also in the process of pair production the gamma-ray energy transferred to kinetic energy of secondary electrons is not  $E_\gamma$  but  $E_\gamma - 2m_0c^2$ , where  $2m_0c^2$  is twice the rest energy of the electron.

Although the total available energy is easily determined, the fraction of this energy actually absorbed within a specific mass of tissue (usually the tissue containing the active material) and, therefore, contributing to the dose is often subject to considerable uncertainty. Gamma rays, except possibly those in the soft x-ray region, are never completely absorbed in the organ from which they originate since the absorption (aside from a geometrical factor of  $1/r^2$ ) is exponential with a half-value thickness greater than the linear dimensions of large organs. The same difficulty affects beta emitters as well. When the dimensions of the organ containing the isotope are comparable to or smaller than the range of beta particles emitted, a considerable fraction of the radiation is only partially absorbed and, consequently, cannot contribute its entire energy to the organ under consideration. Even in very large organs some loss of energy may be sustained due to escape of radiation from active material lying near the surface. At the present time the fraction of available energy contributing to the dose received by a specified mass of tissue can be calculated for only those cases involving the simplest physical and geometrical considerations. In other instances estimates of the dose received can often be made by graphical methods. They may also be made empirically with the aid of phantoms and ionization chamber probes.

**16.2. Units of Dose.** Many units of dose have been proposed in order to circumvent the limitations of the roentgen. Thus far, however, only the roentgen has been accepted by international agreement, while the various proposed units have met with varying degrees of success through common usage. Those units which have found the most frequent use in the United States and Great Britain are described below.

*a. Roentgen.* A dose of one roentgen, received at any point, means

- 1 esu of ion pairs produced per cc air
- $2.083 \times 10^9$  ion pairs produced per cc air
- $1.61 \times 10^{12}$  ion pairs produced per gm air
- $6.77 \times 10^4$  mev absorbed per cc air
- $5.2 \times 10^7$  mev absorbed per gm air
- 83 ergs absorbed per gm air

Air refers to dry air at 0°C, 760 mm Hg. Values of energy absorbed are based on 32.5 ev absorbed to form one ion pair by an electron in air [9,12].

Definition: The roentgen is "that quantity of x- or gamma radiation such that the associated corpuscular emission per 0.001293 gm of air, dry, 0°C, 760 mm Hg produces, in air, ions carrying 1 electrostatic unit of quantity of electricity of either sign" [1].

The roentgen, by its definition, is a partial description of the electromagnetic radiation at a point measured in terms of the ionization produced in air. It is a unit not of intensity, energy, or flux but rather of the time integral of the flux density evaluated according to its ability to ionize air. The relationship that does exist between the energy absorbed in air exposed to one roentgen and energy flux (number of photons per square centimeter  $\times$  energy,  $h\nu$ , per photon) shows a complicated dependence on gamma-ray energy. As shown in Fig. 114, x- or gamma rays of very low energy, corresponding to the energy range in which the photoelectric effect is the principal absorption process, produce the greatest ionization or dose for a given energy flux. Only in the range from 0.08 to about 1.2 mev, where Compton scattering is the principal interaction process, is the roentgen nearly independent of gamma-ray energy for constant flux. In this region 1 r corresponds to approximately 2,800 ergs per cm<sup>2</sup>. In general, however, for a given energy flux the dose in roentgens decreases with increasing gamma-ray energy, at least for energies less than about 30 mev.

An important feature of the definition of the roentgen is its reference to air. One roentgen of gamma rays is the quantity of radiation such that about 83 ergs are absorbed per gram of air, but in substances of different atomic number and density the amount of energy absorbed for this same quantity of radiation will be different. Thus, in soft tissue the energy absorbed per gram per roentgen is about 93 ergs, and in bone it may be several hundred ergs. Despite the great variation in the relative amounts of energy absorbed in different substances, the dose is still 1 r if the same quantity of radiation produces 1 esu of charge of either sign per cubic centimeter of air at the point under consideration. The dose expressed in roentgens is totally independent of the absorbing medium that is exposed and of the amount of energy that the particular medium absorbs.

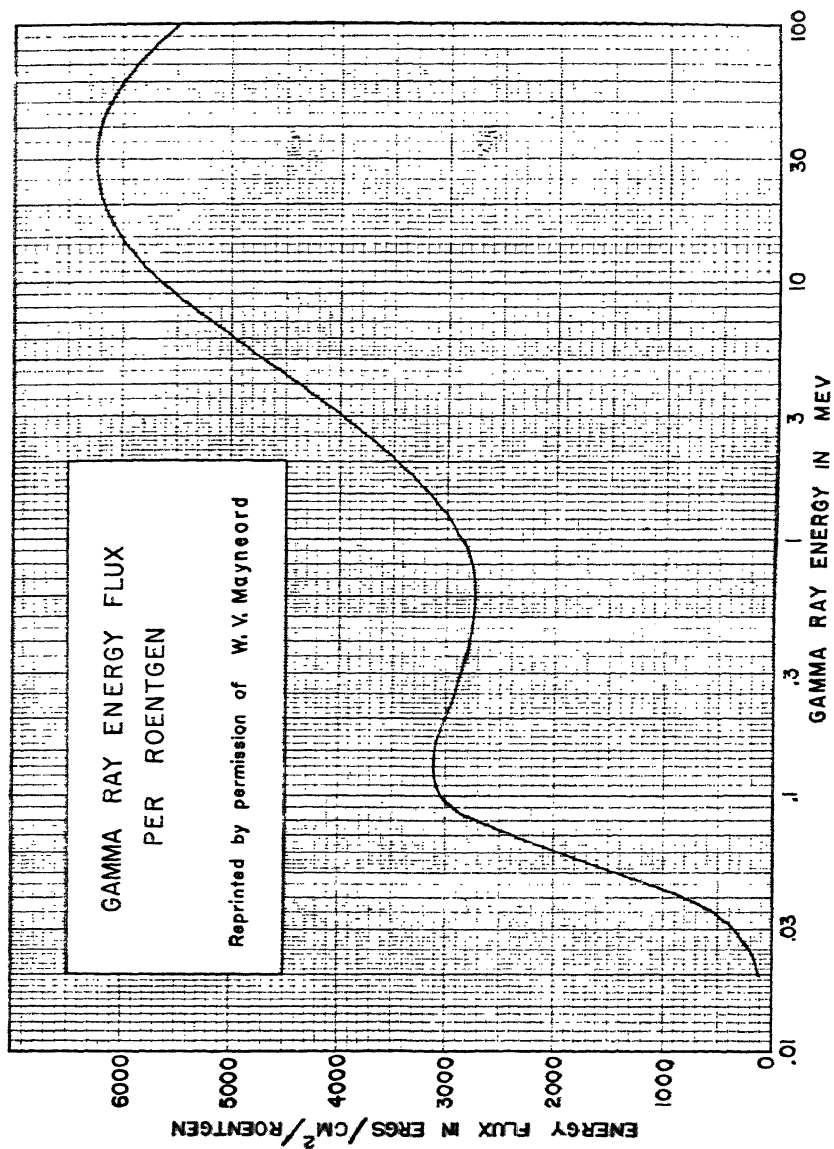


FIG. 114

The roentgen is in some respects a measure of virtual energy absorbed, *i.e.*, the time integral of the energy absorbed if the medium were replaced with air. A dose of 1 r is delivered only when 1 esu of charge is formed per cubic centimeter of air, but this amount of ionization may be produced in any arbitrary interval of time. The time is introduced only in reference to dosage rate expressed in roentgens per unit time. For any given gamma-ray energy the dosage rate is directly proportional to the intensity of radiation.

*b. Roentgen Equivalent Physical (proposed by H. M. Parker [19]).* A dose of one roentgen-equivalent-physical means

$$\begin{aligned} &83 \text{ ergs absorbed per cc tissue} \\ &5.2 \times 10^7 \text{ ev absorbed per cc of tissue} \end{aligned}$$

Definition: "That dose of any ionizing radiation which produces energy absorption of 83 ergs per cubic centimeter of tissue is 1 rep" [19].

The rep is a dose unit applicable to all corpuscular radiations such as beta and alpha particles, protons, and deuterons and is a measure of energy absorbed in tissue exposed to these radiations. Its value is established on the basis of the energy absorbed in air exposed to 1 r, but the rep is not, in general, equal to the energy absorbed per gram of tissue exposed to 1 r. The energy absorbed in tissue exposed to gamma-radiation depends on the atomic composition and density of the tissue as well as on the energy of the radiation, whereas, the rep is always 83 ergs per gram of tissue, independent of tissue composition, type of corpuscular radiation, and energy. In soft tissue, for example, a dose of 1 r corresponds to the absorption of about 93 ergs per gram, whereas this amount of energy absorbed from corpuscular radiation corresponds to a dose of 1.1 rep. If it is known either from computation or experiment that  $E_1$  ergs or  $E_2$  mev of energy are absorbed per gram of tissue, the dose delivered expressed in rep is  $E_1/83$  or  $E_2/5.2 \times 10^7$ , respectively.

The rep is essentially the same unit as the equivalent roentgen (er), used by Marinelli, *et al.* [13], and such units as tissue roentgen and roentgen equivalent proposed by others.

*c. Gram Roentgen (proposed by W. V. Mayneord [12]).* A dose of one gram roentgen means

$$\begin{aligned} &83 \text{ ergs energy absorbed} \\ &5.2 \times 10^7 \text{ ev energy absorbed} \end{aligned}$$

Definition: the gram-roentgen is that amount of gamma-ray energy converted into kinetic energy of secondary electrons which is equal to the energy absorbed by 1 gm of air exposed to 1 roentgen.

The gram roentgen is a unit of energy conversion but without regard to the quantity of tissue in which this amount of energy is absorbed. It is, therefore, a unit of integral dose and should be clearly distinguished from other



dose units as such. It can, however, be expressed as a dose unit energy absorbed per gram of tissue at a point by writing it as a gram roentgen per gram of tissue. From its definition,  $1 \text{ gm r} = 83 \text{ ergs}$ , assuming that the energy required to form one ion pair in air by a secondary electron is 32.5 ev. Like the rep, the gram roentgen represents an amount of energy absorbed independent of tissue composition and is not equal to the energy absorbed per gram of tissue exposed to 1 r. On the other hand, the relation between gram roentgen and rep is exactly  $1 \text{ rep} = 1 \text{ gm r}$  per gram of tissue.

*d. Energy Unit (proposed by L. H. Gray [11]).* A dose of one energy unit means

93 ergs absorbed per gm water (soft tissue)

Definition: The energy unit is that dose delivered to tissue by ionizing radiation such that the energy absorbed per gram of tissue is equal to the energy absorbed per gram of water exposed to 1 r of gamma radiation.

The energy unit is a unit of energy delivered to tissue by any kind of radiation, and its magnitude is chosen so that the energy absorbed in soft tissue is the same whether it is expressed in roentgens or energy units. This equality is not valid however for hard tissue and other substances differing from water (soft tissue) in atomic composition and density.

The relations between eu, rep, and gm r per gm are approximately

$$1 \text{ eu} = 1.1 \text{ rep} = 1.1 \text{ gm r per gm}$$

*e. J Unit [10].*

Definition: "One J has been received at any point in a medium when the ionization which would have been observed in an infinitesimal cavity containing the point is  $1.58 \times 10^{12}$  ion pairs per gram of air enclosed in the cavity."

The J is a unit of dose proposed by the British Committee for Radiological Units (1948) and intended to replace existing dose units. It defines dose in terms of a quantity that is measured directly, namely, the ionization produced in the gas of a cavity ionization chamber (see Sec. 16.1). Because of the properties of the cavity chamber, the J is applicable to all ionizing radiations, electromagnetic and corpuscular, and is valid for all energies of the primary radiation. The numerical value of the J in terms of ion pairs is chosen to correspond to the number of ion pairs formed in water or soft tissue exposed to 1 r. Consequently, 1 J ( $1.58 \times 10^{12}$  ion pairs per gram of air) corresponds to 93 ergs absorbed per gram of air, or alternatively, air exposed to 1 r (1.61 ion pairs per gram of air) of gamma rays not appreciably absorbed photoelectrically corresponds to a dose of 1.02 J.

*f. Roentgen Equivalent Man (Mammal) (proposed by H. M. Parker [19]).* A dose of one roentgen equivalent man has approximately the following significance:

The roentgen is in some respects a measure of virtual energy absorbed, *i.e.*, the time integral of the energy absorbed if the medium were replaced with air. A dose of 1 r is delivered only when 1 esu of charge is formed per cubic centimeter of air, but this amount of ionization may be produced in any arbitrary interval of time. The time is introduced only in reference to dosage rate expressed in roentgens per unit time. For any given gamma-ray energy the dosage rate is directly proportional to the intensity of radiation.

*b. Roentgen Equivalent Physical (proposed by H. M. Parker [19]).* A dose of one roentgen-equivalent-physical means

83 ergs absorbed per cc tissue

$5.2 \times 10^7$  ev absorbed per cc of tissue

Definition: "That dose of any ionizing radiation which produces energy absorption of 83 ergs per cubic centimeter of tissue is 1 rep" [19].

The rep is a dose unit applicable to all corpuscular radiations such as beta and alpha particles, protons, and deuterons and is a measure of energy absorbed in tissue exposed to these radiations. Its value is established on the basis of the energy absorbed in air exposed to 1 r, but the rep is not, in general, equal to the energy absorbed per gram of tissue exposed to 1 r. The energy absorbed in tissue exposed to gamma-radiation depends on the atomic composition and density of the tissue as well as on the energy of the radiation, whereas, the rep is always 83 ergs per gram of tissue, independent of tissue composition, type of corpuscular radiation, and energy. In soft tissue, for example, a dose of 1 r corresponds to the absorption of about 93 ergs per gram, whereas this amount of energy absorbed from corpuscular radiation corresponds to a dose of 1.1 rep. If it is known either from computation or experiment that  $E_1$  ergs or  $E_2$  mev of energy are absorbed per gram of tissue, the dose delivered expressed in rep is  $E_1/83$  or  $E_2/5.2 \times 10^7$ , respectively.

The rep is essentially the same unit as the equivalent roentgen (er), used by Marinelli, *et al.* [13], and such units as tissue roentgen and roentgen equivalent proposed by others.

*c. Gram Roentgen (proposed by W. V. Mayneord [12]).* A dose of one gram roentgen means

83 ergs energy absorbed

$5.2 \times 10^7$  ev energy absorbed

Definition: the gram-roentgen is that amount of gamma-ray energy converted into kinetic energy of secondary electrons which is equal to the energy absorbed by 1 gm of air exposed to 1 roentgen.

The gram roentgen is a unit of energy conversion but without regard to the quantity of tissue in which this amount of energy is absorbed. It is, therefore, a unit of integral dose and should be clearly distinguished from other

dose units as such. It can, however, be expressed as a dose unit—energy absorbed per gram of tissue at a point—by writing it as a gram roentgen per gram of tissue. From its definition,  $1 \text{ gm r} = 83 \text{ ergs}$ , assuming that the energy required to form one ion pair in air by a secondary electron is 32.5 ev. Like the rep, the gram roentgen represents an amount of energy absorbed independent of tissue composition and is not equal to the energy absorbed per gram of tissue exposed to 1 r. On the other hand, the relation between gram roentgen and rep is exactly  $1 \text{ rep} = 1 \text{ gm r}$  per gram of tissue.

*d. Energy Unit (proposed by L. H. Gray [11]).* A dose of one energy unit means

93 ergs absorbed per gm water (soft tissue)

Definition: The energy unit is that dose delivered to tissue by ionizing radiation such that the energy absorbed per gram of tissue is equal to the energy absorbed per gram of water exposed to 1 r of gamma radiation.

The energy unit is a unit of energy delivered to tissue by any kind of radiation, and its magnitude is chosen so that the energy absorbed in soft tissue is the same whether it is expressed in roentgens or energy units. This equality is not valid however for hard tissue and other substances differing from water (soft tissue) in atomic composition and density.

The relations between eu, rep, and gm r per gm are approximately

$$1 \text{ eu} = 1.1 \text{ rep} = 1.1 \text{ gm r per gm}$$

*e. J Unit [10].*

Definition: "One J has been received at any point in a medium when the ionization which would have been observed in an infinitesimal cavity containing the point is  $1.58 \times 10^{12}$  ion pairs per gram of air enclosed in the cavity."

The J is a unit of dose proposed by the British Committee for Radiological Units (1948) and intended to replace existing dose units. It defines dose in terms of a quantity that is measured directly, namely, the ionization produced in the gas of a cavity ionization chamber (see Sec. 16.1). Because of the properties of the cavity chamber, the J is applicable to all ionizing radiations, electromagnetic and corpuscular, and is valid for all energies of the primary radiation. The numerical value of the J in terms of ion pairs is chosen to correspond to the number of ion pairs formed in water or soft tissue exposed to 1 r. Consequently, 1 J ( $1.58 \times 10^{12}$  ion pairs per gram of air) corresponds to 93 ergs absorbed per gram of air, or alternatively, air exposed to 1 r (1.61 ion pairs per gram of air) of gamma rays not appreciably absorbed photoelectrically corresponds to a dose of 1.02 J.

*f. Roentgen Equivalent Man (Mammal) (proposed by H. M. Parker [19]).* A dose of one roentgen equivalent man has approximately the following significance:

$$\begin{aligned}1 \text{ rem} &\simeq 1 \text{ rep for beta particles} \\&\simeq 0.25 \text{ rep for protons, deuterons, and neutrons} \\&\simeq 0.1 \text{ rep for alpha particles}\end{aligned}$$

Definition: The rem is that dose which, delivered to man (or mammal) exposed to any ionizing radiation, is biologically equivalent to the dose of 1 r of x- or gamma radiation (not photoelectrically converted).

The rem was proposed as a practical unit of dose to circumvent the difficulties imposed on other units by the discrepancies in the relative biological effectiveness of various radiations. Hence, the rem is not a measure of energy absorbed or of the ionization produced in tissue; rather, it is a measure of the quantity of radiation that produces certain observable biological effects. The magnitude of the rem in terms of other units is not known with certainty because of the difficulties in evaluating biological effects quantitatively. As yet no wholly satisfactory biological indicator is known since different tissues and organs as well as different species of animals exhibit marked variation in radiation resistance. For this reason the relative biological effectiveness of the same ionization produced, for example, by fast neutrons and gamma rays has been found experimentally to vary from 2 to about 10 in mammals. An approximate value of 4 for the relative effectiveness of neutrons to gamma rays in this instance was derived from the mean lethal neutron dose for rats [5], assuming that the radiation resistance of rats is not significantly different from man.

*g. n-Unit (proposed by R. S. Strong).* A dose of 1 n is approximately

$$\begin{aligned}1 \text{ n} &\simeq 2.5 \text{ rep} \\&\simeq 200 \text{ ergs absorbed per gm of tissue}\end{aligned}$$

Definition: A dose of 1 n is delivered to tissue by fast neutrons when the ionization produced in the Victoreen 100 r thimble chamber equals the ionization produced by 1 r of gamma radiation.

The n-unit provides a convenient means for measuring fast neutron dose with the standard air-wall thimble chamber used in x-ray dosimetry. It does not, however, represent accurately the ionization produced by neutrons in tissue. The dose measured with the thimble chamber represents ionization produced by recoil nuclei in air and from the wall which is airlike in composition, but in tissue the greater percentage of hydrogen increases the conversion of neutron kinetic energy. On the average protons recoil with a greater fraction of the incident neutron's energy than do oxygen and nitrogen atoms. This effect results in tissue ionization 2 to 2.5 times greater than that measured with the air-wall chamber.

**16.3. Calculation of Beta-particle Dose.** The dose delivered to tissue by beta particles (negatrons or positrons) emitted from radioactive isotopes can

usually be estimated by relatively simple methods when the concentration and disposition of the isotope are known. The fact that the range of beta particles in soft tissue is usually only a few millimeters means that the exposed tissue is confined largely to the same regions that contain the active material. For this reason calculations of tissue dose from beta particles, and other low-energy ( $< 15$  mev) corpuscular radiations as well, are essentially estimates of the energy made available by the decay of a certain quantity of active isotope per gram of tissue; the result divided by an appropriate constant permits direct conversion to the desired units of dose. An important factor concerning beta-particle dose should, however, be kept in mind. Although the relatively short range and the consequent localized absorption of beta particles provide a considerable advantage over gamma rays in calculating dose, it cannot be assumed in every instance that a reasonable estimate of dose will be obtained by considering the entire beta energy to be absorbed solely in the tissue or organ containing the isotope. This assumption is valid only when the linear dimensions of the organ are large compared to the range of the particles. If the active material is concentrated near the surface of a large organ or in tissues whose linear dimensions are only a few millimeters, a considerable fraction of the more energetic particles is not completely stopped in the tissue but expends part of its energy outside the region. This is especially true when energetic beta emitters such as  $P^{32}$  (1.7 mev) are taken up in the organs of mice and similar small animals. The computed dose, assuming complete absorption of beta radiation in the organ under consideration, may be in error by a considerable amount. This factor is less important for soft beta emitters, such as  $H^3$ ,  $C^{14}$ , and  $S^{35}$ , since the ranges in tissue are much less than a millimeter and few organs are this small.

The range of beta particles expressed in milligrams per square centimeter is very nearly constant among elements of low atomic weight. It can be assumed, therefore, within the accuracy with which dose calculations are possible, that the range in tissue is essentially the same as in water and aluminum for which the range-energy relations are well established. Note, however, that if the range in a medium is to be expressed in centimeters, the range in water expressed in milligrams per square centimeter must be divided by the density of the medium.

Beta particles in the range of energies encountered from radioactive isotopes (0 to 3 mev) lose energy primarily by ionization. Nevertheless, some energy is also lost by radiation (*Bremsstrahlung*) which in turn is absorbed only in large thicknesses of tissue in the same manner as continuous x-rays. The amount of this radiation is difficult to estimate, but because it is usually small compared to energy loss by ionization, it is disregarded in dosimetry. Positrons present a special problem. Each

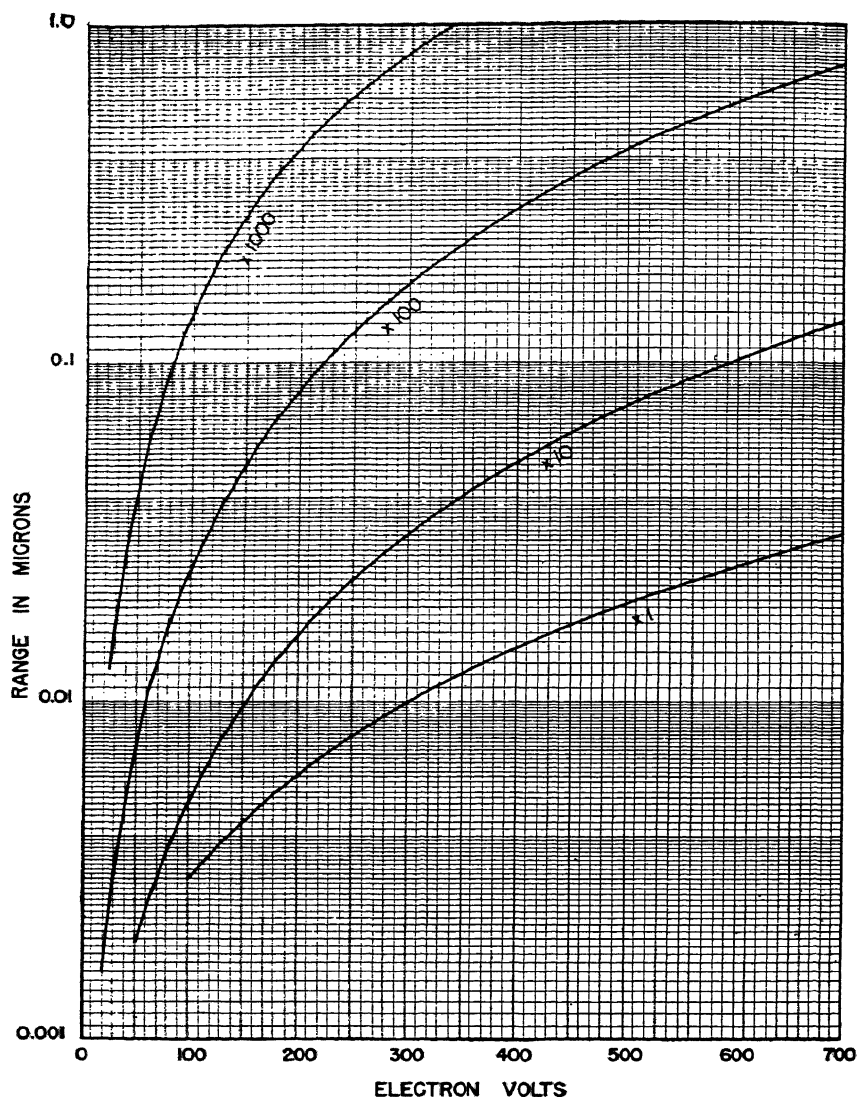


FIG. 115. Range of beta particles in tissue of density-1. [E. J. Williams, *Proc. Roy. Soc. (London)*, **A130**, 310 (1930).]

particle, after losing its kinetic energy to the absorbing medium, undergoes annihilation, producing in the process two gamma rays, each of 0.511 mev ( $m_0c^2$ ). This amount of available energy sometimes cannot be disregarded, and the positron-active isotope may then also be treated as a gamma-ray emitter.

The energy made available per gram of tissue per second by the decay of a beta emitter is given by the product of the number of disintegrations per second per microcurie, the concentration  $u$  of the isotope in microcuries per gram of tissue, and the average energy  $\bar{E}_\beta$  of the beta particles.

$$E_d = 3.7 \times 10^4 u \bar{E}_\beta \quad \text{ev/sec/gm}$$

Similarly, the total energy made available per gram of tissue in the interval of time 0 to  $t$  following administration of active material is

$$E_D = 3.7 \times 10^4 \bar{E}_\beta \int_0^t u \, dt = 3.7 \times 10^4 \bar{E}_\beta U \quad \text{ev/gm}$$

in which  $3.7 \times 10^4 U$  is the number of disintegrations that have occurred in the tissue per gram of tissue during the interval.

The quantities  $u$  and  $U$  depend on the rates of metabolic accumulation and elimination as well as on the rate of radioactive decay. In general, therefore,  $u$  and  $U$  are functions of time and are given in Sec. 16.6 for various cases of biological interest. The simplest possible case may be noted as an illustration. If  $u_0$  microcuries of isotope of long half-life remain fixed in an organ,  $u$  is a constant equal to  $u_0$ . The quantity  $U$  for the interval of time 0 to  $t$  is then

$$U = \frac{u_0 T}{0.693} (1 - e^{-0.693t/T})$$

where  $T$  = decay half-life

The average beta-particle energy  $\bar{E}_\beta$  is not to be confused with maximum energies given in tables of radioactive isotopes. The particles emitted by any isotope vary continuously in energy from zero up to the maximum  $E_{\max}$ . If the beta spectrum is simple and no conversion electrons are present, the average energy in many instances is about  $\bar{E} = E_{\max}/3$ . In general, however, beta spectra are complex; each nucleus decays with emission of one of several possible beta particles for which the maximum energies are different. Furthermore, the presence of strongly converted gamma rays greatly influences the average energy. Under either of these conditions reliable values of  $\bar{E}_\beta$  can be determined only from the observed energy distribution obtained with the beta spectrograph. Values of  $\bar{E}_\beta$  determined by this method for some of the commonly used isotopes are given in Table 6.

With the available energy calculated from the formulas above the dose

received by the tissue in terms of conventional units is easily found as follows:

a. Dosage rate in rep per sec or in gm r per gm per sec

$$d = \frac{E_d}{52.5 \times 10^{12}} = \frac{3.7 \times 10^4}{52.5 \times 10^{12}} u\bar{E}_\beta \quad \begin{array}{l} \text{rep/sec} \\ \text{or gm r/gm/sec} \end{array}$$

where  $52.5 \times 10^{12}$  is the factor for conversion of ev of energy absorbed to rep.

b. Accumulated and total dose in rep

$$D = \frac{E_D}{52.5 \times 10^{12}} = \frac{3.7 \times 10^4}{52.5 \times 10^{12}} U\bar{E}_\beta \quad \begin{array}{l} \text{rep} \\ \text{or gm r/gm} \end{array}$$

c. Integral dosage rate in gm r per sec

$$I.d. = \frac{mE_d}{52.5 \times 10^{12}} \quad \text{gm r/sec}$$

where  $m$  is the weight of the tissue containing active material.

d. Accumulated and total integral dose

$$I.D. = \frac{mE_D}{52.5 \times 10^{12}} \quad \text{gm r}$$

e. Dosage rate in ionization units

$$d = \frac{E_d}{1.58 \times 10^{12} W} \quad \text{J/sec}$$

f. Accumulated and total dose in ionization units

$$D = \frac{E_D}{1.58 \times 10^{12} W} \quad \text{J}$$

the factor  $1.58 \times 10^{12}$  is the number of ion pairs per J, and  $W$  is the average energy required to form one ion pair in the gas of a cavity ionization chamber placed in the medium. For air,  $W = 32.5$  ev per ion pair formed by an electron.

In principle,  $d$ ,  $D$ ,  $I.d.$ , and  $I.D.$  should be multiplied by a fraction  $f$  ( $\leq 1$ ) to take into account the available energy that is not absorbed in the tissue under consideration. If, judging from the linear dimensions of the tissue mass relative to the beta-particle range, it can be assumed that the available decay energy is absorbed almost entirely within the tissue, then  $f \simeq 1$ . If the dimensions of the tissue mass are relatively small, then  $f < 1$  and can be evaluated only by graphical methods [16] or, if the beta-particle absorption coefficient for the substance is known, the beta particles may be treated by the same method used for computing gamma-ray dose (see Sec. 16.5).



The method for calculating beta-particle dose may be illustrated by a simple example. Assume that a colloidal suspension of chromic phosphate containing 2 millicuries of  $P^{32}$  is administered to a dog and that 75 per cent is rapidly accumulated and fixed in the liver which weighs 300 grams. The only factor affecting the dosage rate is then the reduction in  $n$  resulting from radioactive decay. It is assumed that all beta particles are stopped in the liver and that the isotope is uniformly distributed. The initial ( $t = 0$ ) dosage rate is

$$d = \frac{(3.7 \times 10^7)(2 \times 0.75)(6.95 \times 10^5)(60)}{(300)(52.5 \times 10^{12})} = 0.147 \quad \text{rep/min}$$

After 10 days the dosage rate has diminished to

$$d = 0.147e^{-0.693 \times 10/14.5} = 0.0913 \quad \text{rep/min}$$

The accumulated dose delivered to the liver during the first ten days is

$$D = \frac{(0.147)(14.5)(60 \times 24)}{0.693} (1 - e^{-0.693 \times 10/14.5}) = 1,683 \quad \text{rep}$$

Similarly, the total dose (complete decay of the isotope) is

$$D = \frac{(0.147)(14.5)(60 \times 24)}{0.693} = 4,400 \quad \text{rep}$$

The integral dosage rate and accumulated integral dose are obtained in units of gram roentgens by multiplying through by the weight of the liver (300 gm).

**16.4. Absorption of Gamma Rays in Tissue.** From the point of view of dosimetry the absorption of gamma radiation is concerned primarily with conversion of gamma-ray energy to kinetic energy of secondary electrons. While it is true that the energy given to a single secondary electron is not absorbed in the tissue at the point where the photon interacts with the electron, *i.e.*, the electron causes ionization along a path which at high energies may extend a considerable distance, nevertheless, when radiative equilibrium exists between the primary and secondary radiation, the amount of gamma-ray energy converted to kinetic energy determines directly the density of ionization produced at each point in the tissue. Transfer of energy takes place by three distinct physical processes: the photoelectric effect, Compton scattering, and pair formation. Each of these processes depends in a different and complicated way on the gamma-ray energy, and the photoelectric effect and pair formation are, in addition, strongly influenced by the atomic composition of the absorber.

A more detailed description of the interaction of gamma rays with matter is given in Chap. 2, and only those properties important to dosimetry,

essentially the problem of gamma-ray absorption in light elements, are discussed below.

The photoelectric effect is important only at relatively low energies,  $< 0.1$  mev, and in the energy range of soft x-rays ( $< 25,000$  ev) it is almost

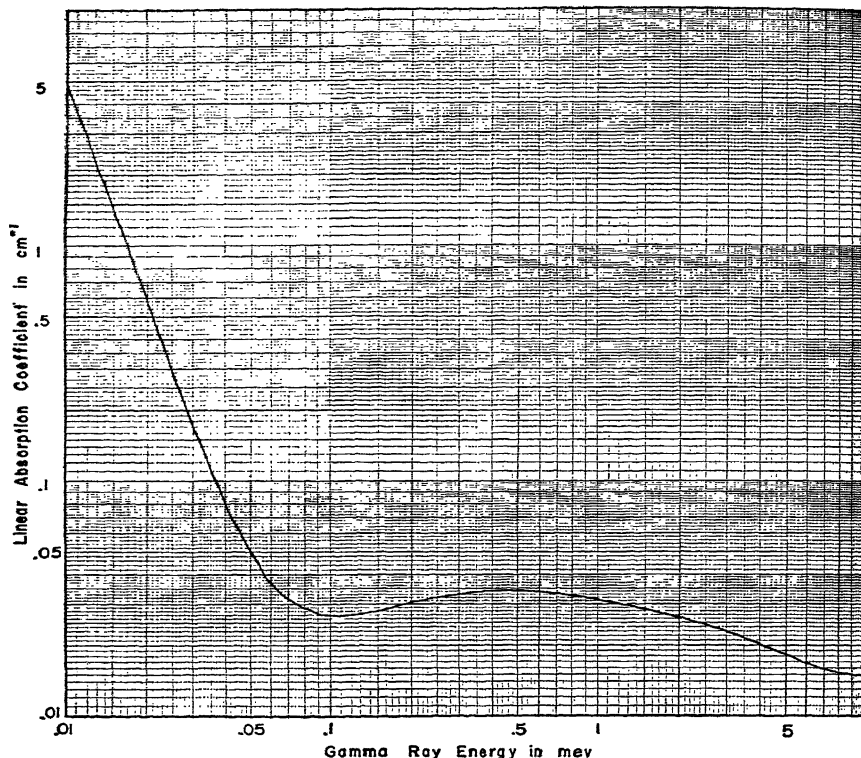


FIG. 116. Linear-absorption coefficient  $\mu_a$  of water as a function of gamma-ray energy in mev. The absorption coefficient includes the photoelectric effect, scattering absorption, and pair production but does not include the contribution from Compton scattering. [From L. D. Marinelli, E. H. Quimby, and G. J. Hine, *Am. J. Roentgenol. Radium Therapy*, **59**, 260 (1948).]

wholly responsible for absorption of electromagnetic radiation in tissue. It involves the complete absorption of a gamma photon by an atomic electron that recoils from the atom with a kinetic energy  $E_e = E_\gamma - I$ , where  $I$  is the ionization potential of the shell, K, L or M, from which the electron is ejected. In light elements composing tissue,  $I$  is usually small compared to  $E_\gamma$ , and for the purposes of dosimetry is neglected. The photoelectric

cross section per electron and per atom and the linear- and mass-absorption coefficients are designated by  $\tau_e$ ,  $\tau_a$ ,  $\tau_l$ , and  $\tau_m$ , respectively.

Compton scattering is usually the most important absorption process for gamma-ray energies  $0.1 < E_\gamma < 3$  mev associated with radioactive isotopes. Each photon that collides with an electron undergoes a deflection from its initial direction and a partial loss of energy, the amount of which is contributed to kinetic energy of the recoil electron. The absorption coefficient  $\sigma_e$ ,  $\sigma_a$ ,  $\sigma_l$ , or  $\sigma_m$  is the sum of a scattering and a scattering-absorption coefficient  $\sigma = {}_s\sigma + {}_a\sigma$ . In nearly all internal-dose problems the coefficient  ${}_a\sigma$  alone should be used since only this term represents the transfer of energy to recoil electrons.

Pair production cannot occur for gamma-ray energies less than 1.02 mev ( $2m_0c^2$ ) and does not contribute appreciably to gamma-ray absorption in tissue except for energies greater than several mev. When pair production occurs, the amount of energy transformed to kinetic energy of recoil electrons is  $E_\gamma - 2m_0c^2$  since the amount of energy  $2m_0c^2$  is required to create the positron and negatron. This amount of energy is subsequently regained in the form of two gamma rays, each of 0.511 mev, on annihilation of the positron. The contribution of annihilation radiation to the dose is greatest for primary gamma-ray energies of 5 to 10 mev, but Mayneord [15] has shown that the effect produced by the annihilation radiation is at most only a few per cent of the dose delivered by the primary radiation and can, therefore, usually be neglected. The absorption coefficients for pair formation are designated by  $\kappa$  with appropriate subscripts  $e$ ,  $a$ ,  $l$ , and  $m$  as for  $\tau$  and  $\sigma$ .

Except in limited energy ranges the absorption of gamma rays must be represented by all three interaction processes. The total absorption coefficient is then  $\mu = \tau + {}_a\sigma + \kappa$ . The units of  $\mu$ ,  $\tau$ ,  $\sigma$ , and  $\kappa$  are largely a matter of convenience, but those commonly used are the cross section per atom  $\mu_a$ , the linear-absorption coefficient  $\mu_l$  in centimeters, and the mass coefficient  $\mu_m$  in square centimeters per gram. These are simply related by

$$\mu_l = \rho\mu_m = \rho N\mu_a$$

where  $\rho$  = density of absorber, gm per cc

$N$  = number of atoms per gm

Although the absorption coefficient  $\mu$  is a complicated function of energy, in tissuelike substances it changes very slowly for energies greater than about 0.1 mev. Nevertheless, the variation in  $\mu$  with energy is sufficiently great so that if the radiation consists of several monoenergetic components with very different energies each component should be considered separately in computing its contribution to tissue dose. The actual dose is then obtained as the sum of the separate contributions. If, however, the energies of the components are greater than 0.1 and are not very different, the com-

ponents can usually be lumped together and an average value of  $\mu$  used without introducing errors greater than the other uncertainties in dose calculations.

The media for which dose calculations are usually made, chiefly air, water, and tissue, or substances of similar atomic composition, are not very different from one another in their effect on gamma-ray absorption. Hydrogen alone is anomalous in its photoelectric effect and scattering. It has an absorption coefficient twice that of other light elements, and the presence of hydrogen in a medium will affect correspondingly the energy absorbed for a given amount of radiation. Thus, the energy absorbed per gram of water (93 ergs) exposed to 1r is 11 per cent greater than that absorbed per gram of air (83 ergs). This effect does not occur in pair production, and consequently hydrogenous substances exposed to very high-energy gamma rays absorb energy as would be expected for an element of unit atomic number.

**16.5. Gamma-ray Dose Calculations.** The calculation of internal dose delivered to tissue by gamma rays emitted from radioactive material distributed within an organ or animal is made extremely difficult by the many physical factors affecting the radiation intensity in various parts of the animal, including the organ containing the active materials. Most important among these is the slow rate of absorption of gamma rays in tissue. The energy made available per disintegration in the form of electromagnetic radiation is readily calculated, but this amount of energy is never completely absorbed in the organ containing the radioactive isotope, and indeed, it is usually not completely absorbed by the entire animal. This is especially true in small laboratory animals. An estimate of dose in this instance, based on complete absorption of gamma-ray energy released by  $\mu$  microcuries of administered radioactive isotope, will be in error by a very considerable factor. The dosage rate at any point in an animal often depends, therefore, on how the active material is distributed throughout the whole body. The distinction between gamma rays and beta particles in this regard is illustrated by considering a spherical mass of tissue containing uniformly distributed active material at a density of  $\mu$  microcuries per gm of tissue. The dose delivered by beta particles under these conditions depends only on  $\mu$ , but that delivered by gamma rays depends upon both  $\mu$  and the total mass in which the isotope is distributed; the greater the mass, the more intense is the gamma radiation at each point.

The dose received at a point  $P$ , fig. 117, due to a small volume  $dV$  of active material of density  $\mu$  microcuries per gm of tissue at  $O$  depends on the geometrical reduction in energy flux over the distance  $R$  and absorption of the gamma rays by intervening tissue. In principle, the dose received per unit time at  $P$  due to active material distributed throughout the volume  $V$  is given by the integral

$$d_i = \int_V \frac{IAu}{R^2} e^{-\mu R} dV \quad \text{r sec}$$

and the accumulated dose received in an interval of time  $T$  is

$$D_i = \int_T d_i dt$$

The subscript  $i$  indicates the dose delivered by one monoenergetic gamma-ray component. It will, however, be dropped with the understanding that the formulas refer only to one component. The factor  $I$  is a constant for conversion to roentgens, assuming that the tissue is uniform in density and composition, and  $A$  is a constant giving the number of gamma rays of a particular energy emitted per microcurie of active substance. If gamma rays of more than one energy are present, the actual dose is the sum of the doses delivered by the components  $d = \sum_i d_i$  and  $D = \sum_i D_i$ .

In practice, evaluation of the dose integral above is possible only after the most extreme geometrical and physical simplifications are introduced. The volume  $V$  can take only the most elementary geometrical forms such as spheres, cylinders, disks, and ellipsoids. These shapes may, however, often be used to approximate a given organ or animal. Formulas for dose calculations with these forms are given in Sec. 16.7. The density  $u(R, t)$  of radiative material, in microcuries per gram of tissue, may sometimes be regarded as a point source, as in the case of radium needles, but usually it is assumed to be constant throughout the tissue within a certain region or organ; nevertheless, because of metabolic processes and radioactive decay, the activity density is, in general, dependent on time. Only in the single instance of a long-lived isotope that remains fixed in the tissue can  $u$  be considered a constant. In other cases, when the rates of accumulation and elimination are known,  $u$  may be estimated with formulas given in Sec. 16.6.

Assuming the active material to be distributed uniformly in a particular region, organ, or entire animal that is essentially constant in tissue density and composition, the dosage rate and total dose at one point due to gamma rays of a particular energy can be computed from the somewhat simpler formulas

$$\begin{aligned} d &= IAu(t)g & \text{r/sec} \\ D &= IAU(t)g & \text{r} \end{aligned}$$

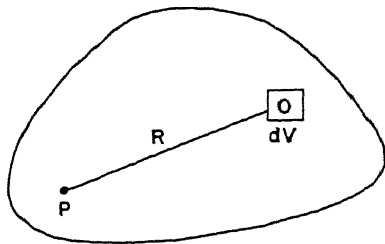


FIG. 117

where  $g$  is the geometrical factor taking into account the volume distribution of active material in some appropriate simple shape which approximates the actual organ. The constants  $I$  and  $A$  are described below.

Integral gamma-ray dose presents a somewhat more difficult problem for which it is necessary to evaluate a double integral or else establish the isodose curves throughout the organ. In units of gram-roentgens the integral dosage rate and total integral dose delivered to a specified volume of tissue are

$$I.d. = 83 I A \rho \int_{V'} \int_V \frac{\mu e^{-\mu R}}{R^2} dV dV' \quad \text{gm r/sec}$$

$$I.D. = 83 I A \rho \int_{V'} \int_V \frac{U e^{-\mu R}}{R^2} dV dV' \quad \text{gm r}$$

or

$$I.d. = 83 I A \mu G \quad \text{gm r/sec}$$

$$I.D. = 83 I A U G \quad \text{gm r}$$

where 83 is a conversion factor (83 ergs absorbed per gram per roentgen) for conversion of energy in ergs to gram roentgens, and  $G$  is the integral geometrical factor (see Sec. 16.7) and  $\rho$  is the tissue density. If the isodose curves have been determined empirically, the integral dosage rate is

$$I.d. = \sum_i 83 \rho d_i \Delta V_i \quad \text{gm r/sec}$$

where  $\rho$  = tissue density

$d_i$  = dosage rate in r per unit time in volume  $\Delta V_i$

$\Delta V_i$  = volume of tissue between isodose surfaces

The number  $A$  of gamma rays of a particular energy emitted per second per microcurie of radioactive isotope depends upon the complexity of the decay scheme of the radioactive isotope. If there is no branching or internal conversion, each isomeric transition occurs once per disintegration or  $A = 3.7 \times 10^4$  gamma rays per second per microcurie of isotope. When decay schemes are complex, branching alone will often make  $A < 3.7 \times 10^4$ . Since gamma radiation may be emitted in isomeric transitions in the nucleus, from annihilation of positrons and as x-rays accompanying K capture and internal conversion, the value of  $A$  must be determined for each component on the basis of the decay scheme.

Isomeric transitions (I.T.) are usually the principal source of gamma rays in most isotopes of biological interest. Assuming that branching and internal conversion (I.C.) occur, the number of gamma quanta emitted per second per microcurie is then

$$A_{\text{I.T.}} = 3.7 \times 10^4 (1 - f) \nu \quad \gamma/\text{microcurie/sec}$$

where  $\nu$  = fraction of disintegrations in which the particular isomeric transition occurs

$f$  = conversion coefficient (defined by  $N_e/(N_\gamma + N_e)$ , where  $N_e$ ,  $N_\gamma$  are numbers of conversion electrons and gamma rays per transition)

The value of  $A$  for annihilation follows in a similar way

$$A_{an} = 2 \times 3.7 \times 10^4 \nu_p$$

where  $\nu_p$  is the fraction of disintegrations in which a positron is emitted. Each positron on annihilation produces two gamma rays, each of 0.511 mev ( $m_e c^2$ ). Nuclei that undergo K capture in a fraction  $\nu_k$  of disintegrations cause the emission of K x-radiation. The number of such x-rays is

$$A_K = 3.7 \times 10^4 \nu_k.$$

The estimation of x-rays associated with internal conversion is often less certain. If only one gamma ray is strongly converted, the value of  $A_{i.c.}$  for all x-rays (K, L, . . .) is  $A_{i.c.} = 3.7 \times 10^4 \nu_{i.c.} f$ , where  $\nu_{i.c.}$  is the fraction of disintegrations in which the isomeric transition occurs. If several gamma ray are converted,  $A = \Sigma A_i$ , summed over all isomeric transitions.

The values of the constants  $\nu$  and  $f$  are given when known in the decay schemes for various isotopes. Unfortunately, accurate values of these quantities are known for only a few isotopes of biological and clinical interest.

The factor  $I$ , as used in the formulas above, is the dose in roentgens received at the surface of a sphere of unit radius due to a single gamma quantum of energy  $E_\gamma$  emitted from the center. For the purpose of its evaluation, consider a single unstable atom placed at the center of a spherical shell of medium with a radius of 1 cm. When the single gamma ray is emitted, the gamma energy converted to kinetic energy of secondary electrons per gram of medium at any point on the unit sphere is

$$E_{K.E.} = \frac{1}{4\pi} [E_\gamma(\tau_m + \sigma\sigma_m) + (E_\gamma - 2m_e c^2)\kappa_m]$$

where  $E_\gamma$  = gamma ray energy, ev

$\tau_m$ ,  $\sigma\sigma_m$ ,  $\kappa_m$  = photoelectric, scattering, and pair production mass-absorption coefficients

Dividing  $E_{K.E.}$  by the energy absorbed per gram of the medium exposed to 1 r, the constant  $I$  is obtained as

$$I = \frac{E_{K.E.}}{nW} \quad \text{r/\gamma ray at cm}$$

where  $n$  = number of ion pairs formed per gm medium per 1 r exposure

$W$  = average energy absorbed to form one ion pair by a secondary electron

If the medium chosen is air,  $n = 1.62 \times 10^{12}$  ion pairs per gram air per

roentgen,  $W = 32.5$  ev and, hence,  $nW = 5.22 \times 10^{13}$  ev or 83 ergs per gram of air per roentgen. Accurate values of  $n$  and  $W$  are not known explicitly for various kinds of tissue, but their product  $nW$  can be determined experimentally. Although the value of  $nW$  for air is sometimes taken

TABLE 37. SPECIFIC GAMMA-RAY DOSES FROM RADIOACTIVE ISOTOPES

Dosage rate in air at 1 cm distance from unfiltered point source of radioactive isotope. Units of  $IA$  are in roentgens per millicurie per hour at 1 cm, milliroentgens per microcurie per hour at 1 cm, or milliroentgens per 10 millicuries per hour at 1 m. Values in parentheses are specific dosage rates due to x-radiation associated with K capture and internal conversion. All other values are for gamma rays from isomeric transitions and from annihilation of positrons. Reported by L. D. Marinelli, E. H. Quimby, and G. J. Hine, *Am. J. Roentgenol. Radium Therapy*, **59**, 260 (1948). Values for Ra are for radium in equilibrium with its decay products and with platinum filtration, reported by W. V. Mayneord, and J. E. Roberts, *Brit. J. Radiology*, **10**, 365 (1937).

Z	El.	A	$IA$ , r/mc/hr at 1 cm	Z	El.	A	$IA$ , r/mc/hr at 1 cm
6	C	11	6.2	29	Cu	61	4.8
						64	1.2
7	N	13	6.2	30	Zn	63	6.9
11	Na	22	13.2			65	3.0 + (5)
		24	19.1	33	As	76	2.2
17	Cl	38	7.6	35	Br	82	15.1
19	K	42	1.95	39	Y	86	14.4 + (3.1)
21	Sc	46	11.4	49	In	111	2.3 + (1.4)
23	V	48	16.3	51	Sb	124	7.9
25	Mn	52	19.5	53	I	128	0.2
		54	4.9 + (11)			130	13.05
		56	9.4			131	2.65
26	Fe	55	(10)	79	Au	198	2.4
		59	6.55		Ra		8.8 (0.25 mm Pt)
27	Co	56	17.95				8.3 (0.5 mm Pt)
		58	5.7 + (7)				6.3 (2.5 mm Pt)
		60	13.5				4.6 (5.0 mm Pt)

to be the same for soft tissue, its true value depends on both the atomic composition of the absorbing medium and the gamma-ray energy. In soft tissue (water), because of the presence of considerable amounts of hydrogen, the value of  $nW$  is found to be about 93 ergs per gm per r.



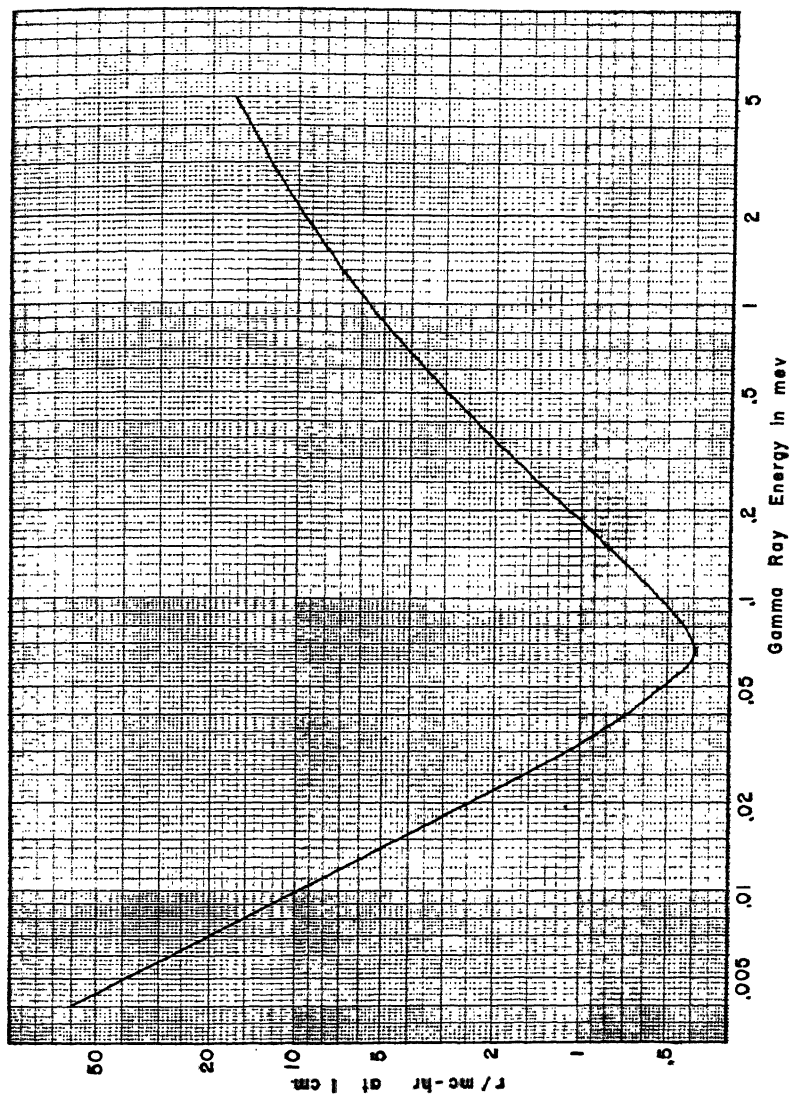


FIG. 118. Dose in roentgens per milliecurie per hour at 1 cm distance in air from point *a* source, as a function of gamma-ray energy in mev. [From L. D. Marinelli, E. H. Quimby, and G. J. Hine, *Am. J. Roentgenol. Radium Therapy*, **59**, 260 (1948).]

The product  $IA$  in the dosage-rate formula has the following important physical significance: it is the dose in roentgens per second at a point 1 cm distant from a point source of 1 microcurie of radioactive isotope for a particular gamma ray (or x-ray) emitted by the isotope. Its value in air calculated by Marinelli, *et al.* [13], as a function of energy is plotted in Fig. 118, assuming one gamma ray per disintegration and a 1-millicurie point source. If the decay scheme is complex or if strong internal conversion occurs, the value of roentgens per millicurie at 1 cm taken from the graph should be multiplied by the factor  $A/3.7 \times 10^7$ , which is the fraction of the disintegrations in which the gamma ray appears.

Experimental values of  $IA$  for air are given in Table 37, but it should be remembered that they represent the dose per hour per millicurie at 1 cm due to all gamma and x-radiation emitted by the substance. In making gamma-ray dose calculations for any isotope the values of  $IA$  given in Table 37 or the graph may be used, or they can be calculated as indicated above.

The foregoing discussion may at first glance appear to place dose calculations generally beyond the scope of anyone but the physicist or radiologist who is thoroughly familiar with the subject. For the most exacting calculations involving complicated distributions of active material and inhomogeneous absorbing material this may perhaps be true, but in many kinds of problems of practical importance even relatively complicated dose calculations can be carried out quickly and with the exercise of nothing more difficult than ordinary arithmetic. Furthermore it is usually unnecessary to consult elaborate tables and charts in order to compute a reasonable approximation of the radiation dose resulting from internal irradiation. The only really essential physical data for this purpose are the energies and percentages of the radiations emitted by the isotope, and the absorption coefficient of tissue for the gamma rays in question. The energies and percentages are given in the table of isotopes (Sec. 7.12) and by the decay scheme (Sec. 7.10) if it is known for the isotope. The absorption coefficient of soft tissue has a value of about 0.03 per cm (or  $\text{cm}^2$  per gm), which is sufficiently accurate for most calculations involving gamma rays with energies from 0.08 to 2.0 mev. The remaining information that is required is more or less physiological and anatomical; at the very least, this must include some information concerning the amount of the isotope taken up in various tissues and the changes in concentration with time.

The comparative simplicity of most dose calculations, as well as the general procedure to follow, is best demonstrated by a typical, although elementary, gamma-ray dose problem worked out in detail. The method described below is easily extended to more involved problems with the aid of formulas discussed in Secs. 16.6 and 16.7. The example to be considered here is the calculation of the gamma-ray dosage rate and the accumulated

dose at the center of the trunk of an average man to whom 0.1 mc of  $\text{Na}^{24}$  is administered.

Tracer investigations have shown [20] that  $\text{Na}^{24}$  given intravenously comes to equilibrium in the interstitial fluids throughout the body within the first 10 min. Following the initial 10-min mixing period, about 27 per cent of the original  $\text{Na}^{24}$  is accumulated in the bones at two different rates for which the corresponding half-times are 10 to 15 min and 1 to 3 hr. Finally, elimination of the  $\text{Na}^{24}$  from the body is found to occur exponentially with a half-time of 12 to 14 days.

The uptake in the bone structure of 27 per cent of the  $\text{Na}^{24}$  does not appreciably alter the gamma-ray dose at various points in the body. The dose may therefore be calculated on the basis of a fixed, uniform distribution of the  $\text{Na}^{24}$ . The justification for this is evident from the fact that the half-value thickness of tissue for the gamma rays from  $\text{Na}^{24}$  is about 25 cm, and consequently within distances of this magnitude the bone structure can also be regarded as uniformly distributed. It should be borne in mind, however, that calculations of the dose delivered by the beta particles to bone and to soft tissue must take into account the uptake of  $\text{Na}^{24}$  in bone.

Finally, the data and approximations required for computing the gamma-ray dosage rate and accumulated dose in this example may be listed in detail:

1. *Point in the body for which the dose is to be determined.* The trunk may be approximated geometrically by a cylinder of tissue 60 cm long and 40 cm in diameter. The point under consideration then lies on the axis midway between the ends of the cylinder and represents the region in the body receiving the maximum gamma-ray dose. The total body weight is assumed to be 60,000 gm.

2. *Effective half-time of  $\text{Na}^{24}$ .* The elimination half-time of  $\text{Na}^{24}$  is long compared to the decay half-life of 14.8 hr, hence only the latter need be considered in the accumulated dose calculation.

3. *Gamma rays.* Two gamma rays are emitted with energies of 1.38 and 2.76 mev in each disintegration (see Sec. 7.10).

4. *Absorption coefficient.* The absorption coefficients for the 1.38 and 2.76 mev gamma rays are about 0.03 and 0.024  $\text{cm}^2$  per gm respectively.

5. *Distribution of  $\text{Na}^{24}$ .* The total quantity of 0.1 mc  $\text{Na}^{24}$  is assumed to be uniformly distributed throughout the cylindrical volume.

The dosage rate at the center of the trunk may now be calculated from the formula  $d = IAug$  for each gamma-ray component and for any time following administration of  $\text{Na}^{24}$ . For the 1.38-mev gamma ray, the factors in the formula takes the following form and values:

$$I = \frac{1}{4\pi} (\text{gamma-ray energy})(\text{absorption coefficient}) \left( \frac{\text{ergs}}{\text{ev}} \right) \left( \frac{\text{roentgen}}{\text{erg in tissue}} \right)$$

$$= \frac{1}{4\pi} \times 1.38 \times 10^6 \times 0.03 \times 1.6 \times 10^{-12} \times \frac{1}{93} = 5.67 \times 10^{-11}$$

$$A = \left( \frac{\text{disintegrations}}{\text{millicurie-sec}} \right) \left( \frac{\text{gamma rays}}{\text{disintegration}} \right) \left( \frac{\text{sec}}{\text{hr}} \right) = 3.7 \times 10^7 \times 1.0 \times 3600 \\ = 1.33 \times 10^{11}$$

$$u = \left( \frac{\text{millicuries}}{\text{gm of tissue}} \right) (\text{decay factor}) = \frac{0.1}{60,000} e^{-0.693t/T} \\ = 1.67 \times 10^{-6} \times e^{-0.693t/T}$$

$g$  = geometrical factor for cylinder (Sec. 16.7) = 200

Thus, the dosage rate for the 1.38-mev gamma ray is

$$d_1 = I A u g = 2.5 \times 10^{-3} \times e^{-0.693t/T} \text{ r per hr.}$$

Repeating the calculation above for the 2.76-mev component yields

$$d_2 = 4.0 \times 10^{-3} \times e^{-0.693t/T} \text{ r per hr.}$$

Finally the actual gamma-ray dosage rate at the center of the trunk for any time  $t$  following injection of  $\text{Na}^{24}$  is

$$d = d_1 + d_2 = 6.5 \times 10^{-3} \times e^{-0.693t/T} \text{ r per hr.}$$

Neglecting the initial mixing period because it is short compared to the half-time of  $\text{Na}^{24}$  in tissue, the dosage rate immediately after injection is just  $6.5 \times 10^{-3}$  r per hr, and after five days it is  $2.4 \times 10^{-5}$  r per hr. It may be noted that the dosage rate produced by the beta particles from  $\text{Na}^{24}$  immediately after injection is  $2.3 \times 10^{-2}$  rep per hr. The change in dosage rate with time is somewhat more complicated for the beta particles because of the accumulation of sodium in bone.

Calculations of the accumulated gamma-ray dose during any prescribed interval of time presents no added difficulty in this simple case since elimination of sodium depends almost entirely on the decay half-life. Integrating the expression for  $d$  above (see Sec. 16.6) the accumulated dose at any time following injection of  $\text{Na}^{24}$  is

$$D = \frac{6.5 \times 10^{-3} T}{0.693} (1 - e^{-0.693t/T}) \quad \text{r}$$

At the end of the first day, for example, the gamma-ray dose received at the center of the trunk is  $D = 9.4 \times 10^{-2}$  r, and at the end of the fifth day it becomes  $D = 0.14$  r.

The precise significance of a dose calculation such as the example worked out above should always be borne clearly in mind. In this instance the meaning of the computed values is quite evident. The dose was calculated for only one point in the body, the point at the center of the trunk where

presumably the maximum gamma-ray dose is received. If the body were actually a cylinder filled with soft tissue of unit density, the calculated dose would be very nearly correct, but since the body contains large cavities and an extensive bone structure consisting largely of minerals, it can be said only that the computed values should be in the correct order of magnitude. The dose received at any other point in the body should be less than at the center, but it is difficult to make such calculations; at the surface of the skin, for example, the gamma-ray dose may be from one-fourth to nearly one-half that at the center. The necessity for uniform distribution of active material is in this case entirely satisfied, at least initially, but in many other instances it is not so certain. Finally, the contribution to the dose made by the beta particles emitted by the isotope must, of course, also be computed. In most cases, especially where small organs are involved, the beta-particle dose will be many times greater than that produced by the gamma rays.

**16.6. Calculation of Radioactivity Density in Tissue.** The dose delivered to tissue or an organ that has taken up radioactive material in one form or another depends directly on the number of atoms that disintegrate while still within the tissue. The estimate of this number is, with a few notable exceptions, complicated by the involved and often uncertain metabolism of the radioactive isotope itself or the substance in which it is incorporated. In most instances metabolic uptake and elimination of active material exhibit rates that are comparable to and often very much smaller than the rate of radioactive decay. Consequently, the number of atoms that decay while in the particular organ under consideration usually is not equal to the number of unstable atoms that pass through it; some atoms decay before entering and many decay after leaving the organ. An estimate can be made, however, when the metabolism of the isotope is known at least superficially, *i.e.*, the gross rates of uptake and elimination regardless of the chemical changes that the isotope undergoes.

To do this it is necessary to determine either empirically or by calculation the activity density  $\mu$  expressed in microcuries per gram of tissue at any instant, and the function  $3.7 \times 10^4 U$ , the number of disintegration per gram of tissue in a given interval of time.

It is to be expected that a mathematical description of metabolic processes will often be impossible or else lead to inconveniently complicated expressions, particularly where second- or higher order chemical reactions are involved. If it is assumed, however, that uptake and elimination can be described by a linear or exponential function of time, the calculations are reasonably tractable and the results are valid for many biological processes and may serve as a first approximation in more involved cases. In addition, it must be assumed that the density of active material is uniform throughout the organ

for which the dose is to be calculated, the total amount of active material in the organ is then just  $mu$  where  $m$  is the tissue mass.

The activity density  $u$  is calculated from the representative, though not general, equation

$$\frac{du}{dt} = f(u, t) - h(u, t)$$

which states that the rate of change of activity equals the rate of uptake minus the rate of elimination. The functions  $f(u, t)$  and  $h(u, t)$  are arbitrary in that they are chosen to describe each case in accordance with appropriate physical and chemical conditions. The function  $U$  is obtained by integration of  $u$  over the interval of time 0 to  $t$ .

$$U = \int_0^t u \, dt \quad \text{microcurie sec/gm}$$

a. The simplest and also a common dose problem is that in which a given quantity  $u_0$  microcuries of isotope is rapidly fixed in a tissue and subsequently is neither accumulated nor eliminated. The activity density after a time  $t$  is

$$u = \frac{u_0}{m} e^{-0.693t/T} \quad \text{microcuries/gm}$$

where  $m$  = mass of tissue, gm

$T$  = decay half-life of isotope

the function  $U$  for the interval of time 0 to  $t$  is then

$$U = \frac{u_0 T}{0.693m} (1 - e^{-0.693t/T}) \quad \text{microcurie sec/gm}$$

When the entire quantity of isotope decays,  $U = u_0 T / 0.693m$ . It may be recalled that  $3.7 \times 10^4 U$  was the number of radioactive atoms originally present (at time  $t = 0$ ) per gram of tissue.

b. Initially no active material is present in the tissue, but for  $t > 0$  it is taken up and fixed in the tissue under consideration at a constant rate of  $b$  microcuries per gm of tissue per second. This represents the behavior, for instance, when active substances are ingested or breathed at constant rates or when taken up from blood or other reservoirs maintained at a constant level of activity. The activity density at any time  $t > 0$  is then

$$u = \frac{bT}{0.693} (1 - e^{-0.693t/T}) \quad \text{microcuries/gm}$$

where  $T$  = decay half-life

The activity in the tissue increases exponentially and approaches after a long time,  $t \gg T$ , the constant level  $bT/0.693$  microcurie per gm. The disintegration function  $U$  for the interval 0 to  $t$  is

$$L = \frac{bT}{0.693} \left[ t - \frac{T}{0.693} (1 - e^{-0.693t/T}) \right] \quad \text{microcurie sec./gm}$$

After a long interval compared to  $T$  the number of atoms that disintegrate in the tissue, and hence the total dose, increases linearly with time.

If instead of being fixed in the tissue the active material is eliminated exponentially with a half-time  $T_2$ , the formulas above are still valid when  $T$  is replaced by  $T_1 T_2 / (T_1 + T_2)$ , where  $T_1$  is now the radioactive decay half-life.

c. In many cases the active material is taken up and fixed in an organ or certain kinds of tissue at a rate proportional to the activity remaining in a reservoir such as blood when the material is given intravenously or perhaps when given orally. Assuming that no activity is present initially in the organ and no elimination occurs, the activity density at time  $t$  following administration of  $u_0$  microcuries is

$$u = \frac{u_0}{0.693m} e^{-0.693t/T_1} (1 - e^{-0.693t/T_2}) \quad \text{microcuries/gm}$$

where  $T_1$  = decay half-life

$T_2$  = uptake half-time

The accumulated activity reaches a maximum value when

$$t_{\max} = \frac{T_2}{0.693} \log \frac{T_2}{T_1 + T_2} \quad \text{sec}$$

The disintegration function  $U$  for the interval 0 to  $t$  following administration is

$$U = \frac{u_0}{m} \left[ \frac{T_1}{0.693} (1 - e^{-0.693t/T_1}) - \frac{T_1 T_2}{0.693(T_1 + T_2)} (1 - e^{-0.693 \left( \frac{T_1 + T_2}{T_1 T_2} \right) t}) \right] \quad \text{microcurie sec./gm}$$

When the entire quantity  $u_0$  decays, the disintegration function becomes

$$U = \frac{u_0}{m} \frac{T_1^2}{0.693(T_1 + T_2)} \quad \text{microcurie sec./gm}$$

This formula, aside from the factor  $3.7 \times 10^4$ , represents the number of disintegrations that actually occur in the organ during a long time compared to  $T_1$ .

d. A more involved case is encountered when part of the administered active substance is taken up in the tissue under consideration at one rate and removed at another rate. Part of the administered substance may also be taken up in other tissues or eliminated directly. All such processes tend to decrease the concentration in the reservoir, and if the removal is exponential

the tissues that take up most activity are those with the shortest uptake half-times. In any one tissue the activity density following administration of  $u_0$  microcuries of substance is

$$u = \frac{u_0}{m} \frac{T_1 T_3}{T_2 (T_1 - T_3)} (e^{-0.693t/T_1} - e^{-0.693t/T_2}) e^{-0.693t/T_3} \quad \text{microcuries/gm}$$

where  $T$  = radioactive decay half-life

$T_1$  = half-time for removal of active substance by all processes from blood, reservoir, site of injection, or precursor

$T_2$  = uptake half-time of activity in given tissue

$T_3$  = elimination half-time for same tissue

If, as it frequently happens, the metabolic half-times are much shorter than the decay half-life, the last factor, containing  $T$ , can be omitted. Under this condition the maximum activity occurs when

$$t_{\max} = \frac{T_1 T_3}{0.693(T_1 - T_3)} \log \frac{T_1}{T_3} \quad \text{sec}$$

The disintegration function for the interval 0 to  $t$  following administration is

$$U = \frac{u_0}{m} \frac{T T_1 T_3}{0.693 T_2 (T_1 - T_3)} \left[ \frac{T_1}{T_1 + T} (e^{-\frac{0.693(T+T_1)}{T T_1}} - 1) - \frac{T_3}{T_3 + T} (e^{-\frac{0.693(T+T_3)}{T T_3}} - 1) \right] \quad \text{microcurie sec/gm}$$

as before, if  $T \gg T_1, T_2, T_3$ , then

$$U = \frac{u_0}{m} \frac{T_1 T_3}{0.693 T_2 (T_1 - T_3)} [T_1 (e^{-0.693t/T_1} - 1) - T_3 (e^{-0.693t/T_3} - 1)] \quad \text{microcurie sec/gm}$$

**16.7. Geometrical Factor.** The dosage rate in roentgens per second at a certain point in an organ or animal due to a known volume distribution of  $u$  microcuries of active material per gram of tissue was shown in Sec. 16.5 to be given by the expression

$$d = I A u(t) g$$

The geometrical factor  $g$  gives the contributions of all parts of the volume distribution of active material to the dose at the point under consideration. Assuming a uniform distribution of radioisotope and a constant tissue density and composition within the volume  $V$ , then

$$g = \int_V \frac{e^{-\mu R}}{R^2} dV \quad \text{cm}$$



where  $\mu$  = linear-absorption coefficient, cm

$R$  = distance of point from volume element  $dV$

In practice the integral leads to excessively complicated expressions for all but the simplest geometrical shapes of the volume. For the same reason the point in the organ or animal for which the dosage rate is computed cannot always be chosen arbitrarily. Furthermore, it is sometimes convenient to neglect absorption in order to simplify the calculations. This is usually justified when the dimensions of the organ are very much smaller than  $1/\mu$ , where  $\mu$  is the linear-absorption coefficient of tissue for a gamma ray of energy  $E_\gamma$ .

Formulas for  $g$  are given below for various geometries that can be used to approximate organs and tissue masses. It is assumed that both the tissue and radioactive substance are uniform in density and composition throughout the volume.

*a. Geometrical Factor for the Center of Sphere.* For a sphere of radius  $a$ , the value of  $g$  at the center is

$$g = \frac{4\pi}{\mu} (1 - e^{-\mu a}) \quad \text{cm}$$

$$\approx 4\pi a \quad \text{cm}$$

*b. Arbitrary Point in Sphere.* Neglecting absorption, the value of  $g$  for any point at a distance  $c$  cm from the center of a sphere of radius  $a$  cm is [14]

$$g = \pi \left( 2a + \frac{a^2 - c^2}{c} \log \frac{a + c}{a - c} \right) \quad \text{cm}$$

It is seen that at the surface  $c = a$ , and  $g$  is just one-half its value at the center.

*c. Disk.* Considering the disk to be flat with radius  $a$  and thickness  $t$ , then neglecting absorption, the value of  $g$  at any point located a distance  $c$  from the plane of the disk and a distance  $b$  from its axis is [15]

$$g = \pi t \log \frac{a^2 - b^2 + c^2 + \sqrt{(a^2 - b^2 + c^2)^2 + 4a^2b^2}}{2c^2} \quad \text{cm}$$

*d. Cylinder.* The value of  $g$  at the mid-point of the axis of a cylinder of length  $2L$  and radius  $R$  is approximately [13]

$$g = 2\pi \left( L \log \left( 1 + \frac{R^2}{L^2} \right) + 2R \tan^{-1} \left( \frac{L}{R} \right) - \mu \left\{ L \sqrt{R^2 + L^2} \right. \right. \\ \left. \left. - L^2 + R^2 [\log (L + \sqrt{R^2 + L^2}) - \log R] \right\} \right) \quad \text{cm}$$

*e. Graphical evaluation of  $g$*  is possible for any arbitrary point whether inside or outside the tissue mass containing active material and may be

applied as well to irregularly shaped masses. The entire tissue volume is divided (graphically) into small volume elements  $\Delta V_i$  whose dimensions are very small compared to  $1/\mu$ . For an arbitrarily chosen point  $P$  at which the dose is to be determined, the value of  $g$  is computed by a summation of the contributions from all  $\Delta V_i$ ; thus

$$g = \sum_{i=1}^s \frac{e^{-\mu R_i} \Delta V_i}{R_i^2} \quad \text{cm} \quad (1)$$

where  $R_i$  is the distance from the center of  $\Delta V_i$  to the point  $P$ .

The calculations are facilitated by taking as the first term of the sum the largest sphere with  $P$  as its center which lies entirely within the tissue; then

$$g = \frac{4\pi}{\mu} (1 - e^{-\mu a}) + \sum_{i=1}^s \frac{e^{-\mu R_i} \Delta V_i}{R_i^2} \quad \text{cm} \quad (2)$$

where  $a$  is the radius of the sphere and the sum is now taken over the volume divisions lying outside the sphere but in the tissue. Further simplification is possible if the organ permits convenient division into concentric spherical shells of thickness  $\Delta R_i$  surrounding the first solid sphere. These shells will not lie entirely within the tissue, but by estimating the fraction  $f_i$  of each shell which does lie within the organ, the value of  $g$  is then computed from

$$g = \frac{4\pi}{\mu} (1 - e^{-\mu a}) + \sum_{i=1}^s e^{-\mu R_i} f_i \Delta R_i \quad \text{cm} \quad (3)$$

The fraction  $f_i$  is, of course, determined by estimating the surface area of the shell bounded by the organ and dividing by  $4\pi R_i^2$ , where  $R_i$  is the radius to the mid-thickness of the shell.

The determination of integral dose or total energy absorbed by the tissue presents a more difficult problem in that the geometrical factor  $G$  which must be calculated is given by a double integral

$$G = \int_{V'} \int_V \frac{e^{-\mu R}}{R^2} dV dV' \quad \text{cm}^4 \quad (4)$$

where  $V$  is the volume containing uniformly distributed activity,  $V'$  is the volume for which the integral dose is to be determined, and  $R$  is the distance between the volume elements  $dV$  and  $dV'$ . The volumes  $V$  and  $V'$  need not be identical. Although  $V$  is determined, in principle, uniquely by the distribution of the active isotope, the volume  $V'$  may be chosen arbitrarily provided that the factor  $e^{-\mu R}$  is retained. This integral does not reduce to

simple integrable form except for the sphere when absorption is disregarded; then  $G = 4\pi^2 a^4$ , where  $a$  is the radius of the sphere containing the radioactive material and for which the integral dose is determined. In most other cases the evaluation of  $G$  is possible only by numerical integration. In principle,  $G$  can be evaluated by a double summation similar to that for  $g$

$$G = \sum_{j=1}^r \sum_{i=1}^s \frac{e^{-\mu R_{ij}} \Delta V_i \Delta V_j}{R_{ij}^2} \quad \text{cm}^4$$

in which the sum  $i = 1$  to  $s$  is taken over all volume elements  $\Delta V_i$  into which the exposed volume is divided and the sum  $j = 1$  to  $r$  is taken over volume elements  $\Delta V_j$  containing active material.  $R_{ij}$  is the distance from  $\Delta V_i$  to  $\Delta V_j$ . It is apparent that the labor involved in evaluating this double sum will often be prohibitive.

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## CHAPTER 17

# THE PREPARATION OF THIN FILMS OF RADIOACTIVE ELEMENTS BY ELECTROLYSIS

RAYBURN W. DUNN

**17.1. General Considerations.** Although many methods have been proposed for the preparation of thin films that may be used for radioactivity measurements, probably the most convenient and satisfactory is the electrolytic. Under properly controlled conditions it is possible either to electroplate or to electrodeposit many of the elements quantitatively from solutions of their salts, and the resulting film can be made quite uniform in thickness. If the apparatus is correctly designed, the area that this film covers will be both accurately fixed and reproducible.

With certain minor modifications, the electrolytic methods generally employed for quantitative chemical analysis are readily applied to radioassay. For the most part, these methods are for electroplating rather than electrodepositing. The term "electroplating" is usually taken to mean the formation, by electrolysis, of a closely adhering metallic film. "Electrodeposition," on the other hand, may have this same meaning, or it may refer to the electrolytic formation of a nonmetallic layer, closely adherent or not, on the electrode. It is in the latter sense that it is employed here.

Among the elements that have been successfully electroplated from aqueous solution are copper, silver, gold, zinc, cadmium, mercury, tin, lead, antimony, bismuth, chromium, iron, cobalt, nickel, and platinum. Aluminum and several other metals that are not reducible in aqueous solution may be plated from organic solvents. In the group of electrodeposited elements are, among others, uranium, oxygen, and fluorine as  $\text{UO}_2$  and  $\text{UF}_4$ , and thorium, iron, carbon, and nitrogen, as  $\text{ThFe}(\text{CN})_6$ . Quantitative procedures for electrolyzing most of these elements are well known, although only a few have as yet found use in radiochemistry.

In general the methods for commercial electroplating cannot be successfully applied in analytical work, although the chemical principles and reactions of both procedures may be identical. This is essentially true because the conditions of temperature, ionic species and concentration, current and voltage required for quality plating do not necessarily satisfy the requirements for quantitative recovery. It is advantageous, however, to obtain a smooth, shiny film whenever possible. Rough films are likely to be porous and may occlude extraneous salts from the electrolysis solution.

Colloids are frequently used to improve the electroplate. But no specific rules can be given to govern their use.

**17.2. Apparatus.** Many types of electrolysis cells and stirrers have been designed and used by the various groups that are investigating the field of radioassay. Unfortunately, many of these developments, like those of the chemical procedures, have not found their way into the published literature.

The simplest electrolysis apparatus will have a rotating anode of platinum or platinum-10 per cent iridium wire. The wire is usually coiled in a flat spiral and serves both as a conductor and a stirrer. A platinum dish contains the electrolysis solution and serves as the cathode. Alternatively, cathodes consisting of thin disks are probably the most convenient for both gravimetric and radiometric measurements. Cathodes of 1 or  $1\frac{1}{2}$  in. in diameter are most frequently employed. These vary in thickness from 0.0005 to 0.005 in. depending upon the cost of material, the ease of handling, and the effect of background scattering on the counting rate.

A glass cylinder and gasket, each having the same inside diameter and an outside diameter of either 1 or  $1\frac{1}{2}$  in., generally constitute the body of the cell. The base is constructed so that either it serves for the cathode connection itself, or a separate connection is incorporated in it. Finally some means must be provided to hold the glass tightly against the base with the gasket and cathode in between.

**17.3. Anodes.** Platinum wire or gauze anodes have been almost universally used for laboratory electrochemical reactions, principally because they are very passive. Platinum has some limitations, however, of which cost is not the least. Electrolysis of acid chloride solutions produces free chlorine at the anode, and in the presence of this gas, platinum loses its passivity to some extent and small but appreciable amounts are dissolved. This reaction increases with hydrochloric acid concentration, temperature, and current density but is minimized if the platinum is alloyed with 10 per cent iridium.

Similar attack occurs if electrolysis takes place in an alkaline solution containing free ammonia. This attack will not be very great if the current density and ammonium hydroxide concentration can be kept low; however, it may account for cathode deposits of 0.1 to 0.2 mg if lengthy electrolyses are carried out.

Substitutes for platinum have not proved too satisfactory for quantitative analytical work since most are attacked in either acid or alkaline solution. If the cathode film need not be weighed, alloys of the 18-8 variety of stainless steel sometimes may be used; carbon or graphite may be employed if anodic oxygen is not formed and if the rods are not appreciably porous. Tantalum cannot be used because it forms an oxide coating that prevents the flow of current from solution to anode, although the reverse process occurs with ease.

**17.4. Cathodes.** The selection of a satisfactory metal for electroplating or electrodeposition depends largely upon whether gravimetric as well as radio-metric assay determinations are to be made. If the cathode film is to be weighed, it is best to employ platinum disks; if not, copper, iron, etc., may be used. Furthermore if the film is to be ignited or calcined, platinum is to be preferred; it must be used if both heating and weighing are necessary or if the film is later to be recovered quantitatively in a pure state. In most cases gold may be substituted for platinum.

It is generally assumed that cathodes of gold or platinum will be cleaned and reused. Under some conditions it may be impossible to remove completely the radioactive film, in which case the disks must be discarded, or the activity of each recleaned disk must be recorded before the next use. When these alternatives are either impractical or uneconomical, gold-plated copper disks may be substituted. A commercial plate 0.0001 in. is usually sufficiently thick and is relatively inexpensive. After use, the plated gold may be reclaimed, although it is safer to discard the disks rather than to reuse the recovered gold since it may contain contaminating radioactivities.

Although most of the elements that can be electroplated will plate out on and adhere to copper, platinum, and gold, there is no hard and fast rule that can be used to determine exactly what results may be obtained with each element and each cathode. Both electroplating and electrodeposition are greatly influenced by the character of the cathode surface, as well as by the composition of the electrolysis solution. Cathodes of the same material will vary from lot to lot and even from front to back. For these reasons, duplicate determinations will frequently produce films greatly different in appearance and possibly in composition or crystal structure. Presumably identical electrolysis solutions and cathodes will frequently show wide variation in gas production, even though the currents and anode potentials are the same.

The formation of satisfactory films by electrodeposition is more critically influenced by the character of the cathode surface and solution composition than is such formation by electroplating. The greatest problem in electrodeposition is to obtain a film that will adhere to the electrode, and it is for this reason that etching of the surface may be advantageous. Inasmuch as no definite instructions can be given in every case regarding the selection of a satisfactory cathode, it is often necessary to try each metal in turn, starting with platinum or copper.

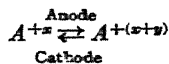
Since the cathode surface is so important to satisfactory film formation, thorough cleaning of disks cannot be overemphasized. Surface impurities result from rolling, handling, dust, oxidation, and attack by moist air containing small amounts of such gases as sulfur dioxide, hydrogen sulfide, hydrochloric acid, and nitric acid which are not usually excluded from the

air in the laboratory. The greasy coating due to handling should be removed before the oxide and salt coating, unless removal of both can be accomplished at the same time. The noble metals, gold and platinum, are readily cleaned by immersion in warm concentrated sulfuric acid-dichromate cleaning solution, and, if desired, this treatment may be followed by boiling in either concentrated nitric or hydrochloric acid.

Copper and iron may be degreased with organic solvents, or by immersing for a very short period in the dichromate cleaning solution, followed by washing with hot water, or they may be cleaned with fine sandpaper. After any of these procedures, immersing in cold dilute hydrochloric or nitric acid will complete the cleaning. Nitric acid will attack and etch the copper surface faster than hydrochloric, while the reverse is true with iron, and advantage may be taken of these rate differences to effect cleaning with or without appreciable etching. As pointed out above, it may sometimes be advantageous to etch the cathode during cleaning, using aqua regia or sand-blasting if necessary. Rinsing the disks in acetone before drying is particularly recommended for cathodes that are easily oxidized.

**17.5. Electrodeposition.** The electrodeposition process involves not one but two different chemical reactions; namely, oxidation or reduction and, concurrently, precipitation of an insoluble salt on the electrode. Electrodeposition, unlike electroplating, can occur on either the anode or the cathode. Anodic depositions, although reported in the literature, have not as yet been successfully applied to the problems of radiometry. Anodic films of lead sulfate and oxide, and of manganese oxides, for example, have been produced, but procedures for quantitative recovery by this means have not been worked out.

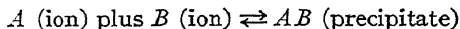
The underlying principle upon which electrodeposition is based can be explained as follows: A cation  $A$ , which can exist in solution in either of two or more valence states, is oxidized at the anode and reduced at the cathode. An anion  $B$ , also in solution, but neither electrolytically oxidizable nor reducible, forms an insoluble compound either with the reduced cation but not the oxidized, or with the oxidized but not the reduced. If the reduced cation deposits with  $B$ , then cathodic deposition is obtained, and by the converse process, anodic deposition may be obtained. These may be represented as follows:



which are the cation electrode reactions, where  $x$  has a value between 1 and 3 and the quantity  $(x + y)$  has a value between 2 and 5. If the anion  $B$  has a valence of  $-z$ , it will form two salts with  $A$ , namely,  $A_zB_z$  and  $A_zB_{(x+y)}$ . For electrode film formation, one of these must be insoluble in the electrolysis

solution. Of necessity the cation must be introduced into the electrolysis solution in such a valence state that it will not form this precipitate in the solution itself.

Neglecting valences and electrolysis reactions for the moment, one other consideration is important, namely, the independent role that  $B$  plays in forming and maintaining the precipitated film. The reaction that occurs may be represented as



The equilibrium constant for this reaction,  $K_{ab}$ , equals the product of the concentrations of  $A$  and  $B$ ; the value of each of the latter is squared or cubed, etc., depending upon whether one, two, or more ions are concerned in forming the precipitated molecule. Therefore the greater the concentration of  $B$  in solution, the smaller will be the concentration of  $A$  when equilibrium is reached and, thus, the more quantitative the recovery.

The electrodeposited film, however, is not only slightly soluble in water, but is also usually dissolved by acid or base, or by complex ion formation. If the rate of solution by any one reaction or combination of these reactions is faster than the rate of valence change and precipitation, no electrode film will form. A rather delicate balance of these reaction rates is usually necessary if quantitative results are to be achieved and such side reactions must be eliminated or minimized.

**17.6. The Electrolysis Current and Voltage.** A convenient power supply for electrochemical reactions is the ordinary 6-volt storage battery. Voltages above 12 are not usually necessary, and a parallel or series-parallel arrangement of two or four batteries will supply as much power as will, in general, be needed for routine analytical work. Electrolyses from nonaqueous solution, however, may require much higher voltages, in which case a vacuum-tube d-c power supply will be much more convenient than either batteries or a generator.

In addition to the voltage, three other independent variables will influence the electrolysis current, namely, the distance between anode and cathode, the resistance of the electrolysis solution, and the electrode-solution boundary resistances. The electrolysis-solution resistance, in turn, is dependent upon the ion concentration and upon the temperature of the solution; increasing either one decreases the resistance.

The desired electrochemical reactions occur only at the boundaries joining the electrodes and the electrolysis solution; therefore the potential drop across each of these boundaries is of considerable importance. These electrode voltage drops determine which reactions will occur, and the current that flows determines the reaction rates. If either of the boundary resist-



ances or the resistance of the solution is too high, little or no current will pass through the cell, and consequently the reaction rates will be negligible.

The minimum electrode reaction potentials are determined by (1) the back emf, which is the potential with which the particular reactions tend to reverse themselves; (2) the "passivity" of the electrodes; and (3) the "polarization" within the boundary. The last-named phenomenon results, primarily, from localized concentration changes of the electrolysis solution; passivity is due to electrode surface conditions for which discussions will be found in standard texts. An electrode becomes polarized owing to the progressive removal of one or more ions, by reduction or oxidation, within the boundary; equilibrium is regained or maintained by rapid stirring of the solution. The net result of the above three factors is to establish a minimum potential below which little or no current will flow and no reactions will occur and above which a rapid increase in current is obtained.

The polarization of an electrode, which is required to produce a given irreversible reaction under given conditions, is known as the overvoltage of this reaction. Overvoltage values for the same reaction vary considerably, depending upon the electrode material and surface condition. Thus the hydrogen overvoltage for a platinized-platinum cathode is lower than for smooth platinum, which, in turn, is lower than for mercury, etc.

The importance of hydrogen production at the cathode may be illustrated by the following example: If the electrode emf for the reaction or reduction of *A* is 0.5 volt and the hydrogen overvoltage is 1.0 volt, reaction *A* will begin when the potential slightly exceeds 0.5 volt and will continue with little or no hydrogen evolution until the potential reaches 1.0 volt. At this point hydrogen gas begins to form and, as the voltage increases, both hydrogen formation and reduction of *A* occur together. The reduction of hydrogen at the cathode is accompanied by the formation of hydroxyl ions. These in turn may exercise considerable influence on the reduction of *A* or upon the character of the reduced film. If this influence is deleterious, then voltages below the hydrogen overvoltage should be employed. On the other hand, increased alkalinity may be helpful; furthermore the presence of hydrogen gas within the body of the solution may cause reductions that will aid in the quantitative recovery of *A*.

When the hydrogen overvoltage is much lower than the voltage necessary for the reaction *A* and the applied voltage is between the two, then no reduction of *A* will occur and only hydrogen gas will be formed. To overcome this, a different cathode material with a higher overvoltage may be used or the voltage may be increased to the point where reaction *A* begins to occur, whereupon the cathode becomes coated and the hydrogen overvoltage increases to a value that is characteristic of a cathode made of metal *A*. Of

course, if the hydrogen overvoltage of the latter metal is lower than the voltage necessary for its plating, then the reduction rate may be very slow and may not occur quantitatively within a finite time.

Oxygen overvoltages and the formation or lack of formation of oxygen gas and hydrogen ions should be considered in exactly the same relation, but with respect to the anode. However, since most of the electrolysis work with radioactive materials will be concerned with cathodic film formation, anodes with low oxygen overvoltages should be employed; this condition is adequately met in platinum.

# CHAPTER 18

## TREATMENT OF BIOLOGICAL TISSUES FOR RECOVERY OF RADIOACTIVE ELEMENTS

RAYBURN W. DUNN

**18.1. Introduction.** Methods for ashing plant and animal tissues have been described by many workers. Descriptions of satisfactory ashing methods are usually included with the corresponding analytical procedures, frequently with only slight variations from previously reported work. There are, therefore, very nearly as many ashing techniques in biological work as there are methods for the estimation of inorganic elements. Since it is doubtful whether any one procedure can be said to be superior to the rest, only a general description of ashing techniques will be given. It should be possible to devise, from the information given, a method that will meet the specific requirements of a particular problem.

A glance at the two tables copied from Hawk will show the relative amounts of inorganic elements that are to be found in human-tissue ash [1] and in blood [2].

TABLE 38. INORGANIC CONSTITUENTS OF HUMAN BLOOD

Constituent	Normal range, mg. 100 ml
Chlorides, as NaCl.....	450-500
Sulfates, inorganic, as S (serum).....	0.9-1.1
Phosphorus, inorganic, as P (plasma).....	3-4
Iron, as Fe.....	52
Copper.....	0.05-0.25
Calcium (serum).....	9.0-11.5
Magnesium (serum).....	1-3
Sodium (serum).....	330
Potassium (serum).....	16-22
Iodine (micrograms per 100 ml).....	8-15

In selecting ashing and purification procedures, the normal presence of these elements must be taken into consideration. The concentrations of calcium, magnesium, sodium, potassium, phosphorus, chlorine, sulfur, etc., in the ash will greatly influence their own recovery and that of other elements present in trace amounts. These concentrations will also dictate the course to be followed in the subsequent analytical procedures.

TABLE 39. INORGANIC CONSTITUENTS OF HUMAN TISSUE

Element	Per cent	Approximate amount, in grams, in a 70-kg Man
Oxygen....	65.0	45,500
Carbon.....	18.0	12,600
Hydrogen..	10.0	7,000
Nitrogen...	3.0	2,100
Calcium....	1.5	1,050
Phosphorus	1.0	700
Potassium..	0.35	245
Sulfur.....	0.25	175
Sodium....	0.15	105
Chlorine...	0.15	105
Magnesium..	0.05	35
Iron .....	0.004	3
Manganese..	0.0003	0.2
Copper.....	0.0002	0.1
Iodine.....	0.00004	0.03

**18.2. Dry Ashing.** Ashing at 500 to 800°C is generally carried out in platinum, porcelain, Vycor, or nickel crucibles, although in general, platinum is to be preferred. It cannot be employed, however, if fluxes are used that contain such materials as potassium acid sulfate, sodium peroxide or hydroxide, excessive amounts of potassium nitrate, or any compounds likely to liberate free chlorine. Such fluxes are not usually encountered in biological work, but if they are needed, it is advisable to use porcelain or Vycor, provided that etching by the alkali is not objectionable. Admixtures of such materials as magnesium acetate and calcium hydroxide will not result in attack on platinum. If the dissolution of ash requires the use of aqua regia, porcelain or Vycor must be used, whereas platinum is indicated when hydrofluoric acid is employed.

One of the most difficult problems encountered when porcelain is used is the fusion of the inorganic material with the crucible; this is a particularly undesirable characteristic of some animal tissues. Although the loss of a small portion of the sample due to crucible fusion may not appreciably affect the analytical results, it may preclude the subsequent use of this crucible. The danger of cross contamination due to residual activity in laboratory glassware must be kept in mind at all times; crucibles showing unremovable residual activity should, therefore, be discarded. Furthermore, if fusion occurs, recovery of the ashed material is not only difficult but time-consuming. Vycor and platinum are more satisfactory under these conditions, although from an economic standpoint it may be less expensive to

use porcelain and to discard the crucibles if appreciable activity remains in them.

For dry ashing the muffle furnace which has heating elements at the sides as well as top and bottom is much to be preferred to gas burners. Not only is the ignition more convenient, but it is also more uniform. The muffle furnace has the disadvantage, however, that when completely filled the available oxygen inside is rapidly exhausted. When this condition is reached, the combustible gases produced will leave the furnace as a tarry and soot-laden smoke; the latter must usually be removed by forced ventilation. To avoid this and to aid in combustion, oxygen may be introduced into the furnace through the thermocouple housing, using a quartz tube. When this procedure is followed, the heating units must be reversed if possible so that the coils do not face toward the inside of the furnace, and furthermore the stream of oxygen must not be directed onto the thermocouple itself.

The general ashing procedure consists of air-drying the weighed sample at a temperature of 100°C or slightly less followed by ignition at 500 to 800°C for a minimum of 2 hr. Samples that have been ground or macerated may be dried at reduced pressures; 100 mm or less is recommended for grain and stock feeds, meat meal, plant tissues, etc. [3]. Caution must be exercised in drying whole tissues, such as liver, since they tend to swell at elevated temperatures or reduced pressures or both. Temperatures above 100°C are permissible for removing the last portions of water and volatile oils. Drying by means of overhead radiant heating has been found to be particularly useful with nearly all types of biological tissues as well as with solutions of both organic and inorganic compounds.

In many cases the material to be ashed is already dry enough to place in the muffle furnace. Ground grains, bone, and the woody portions of plants, for instance, seldom need to be dried before ignition.

When possible cross contamination or contamination of the muffle furnace is likely to influence the results, it is best to place the samples in a cold furnace and to allow the temperature to rise gradually to the value desired. Rapid ashing produces volatile gases at rates that are sufficient to carry small amounts of the material out of the crucible. Thus radioactive deposits will collect on the walls and floor of the furnace as well as in the adjacent crucibles.

The duration of ashing and the temperature must be considered in relation to the volatility of certain compounds. For example, some iron generally will be lost as ferric chloride, phosphorus as phosphoric acid, sulfur as sulfur di- and trioxide, fluorine as hydrogen fluoride or silicon tetrafluoride, and chlorine as hydrogen chloride. Because of the difficulty of completely mixing samples with additives that are designed to prevent these losses, 100 per cent recovery of some elements is impossible. From a quantitative standpoint these losses may be inconsequential, but they must be considered

in light of the possibility of serious radioactive contamination. Wherever feasible, therefore, wet ashing should be considered if the radioactive element is likely to be volatilized at muffle-furnace temperatures.

For radioactive tracer studies, ashing to constant weight is not important. The aim is solely to decompose the tissue to the point where organic compounds, which are likely to interfere in the subsequent analysis, are completely decomposed. Ashing beyond this point, to oxidize the last traces of carbon, is unnecessary and time-wasting. The use of perchloric acid after such incomplete ashing should not only be safe but should also facilitate the dissolution of the inorganic salts. The loss of important elements from the tissue ash might very well be eliminated by this less vigorous procedure; compounds difficult to dissolve should not be produced so frequently.

Aids to ashing have been employed with considerable success. Calcium nitrate, magnesium acetate, and mixtures of calcium and aluminum nitrates may be advantageously used. These salts act not only to supply oxygen, but they also increase the bulk of the ash and render it more amenable to dissolution. The use of these aids is indicated with samples producing little or no ash of their own, *e.g.*, fatty tissues and carbohydrates. Magnesium acetate in alcohol is particularly recommended for use with such finely ground material as flour [4]. Fusion of material with the crucible is practically eliminated when ash aids are employed. Etching of the glaze on porcelain is not completely prevented, however, and chlorine attack on platinum, due to oxidation of chloride by nitrate, is likely to occur.

**18.3. Wet Ashing.** Less drastic than furnace ignition but more satisfactory in many respects are the procedures for oxidation of organic material in solution. Wet digestion may require more attention and will frequently be slower than dry ashing, but on the whole these disadvantages are more than offset by the superior results obtained when the quantitative recovery of certain elements is desired. The choice between wet and dry ashing is based, therefore, upon consideration of the likelihood of loss by volatilization at higher temperatures versus the inconvenience in increased time and labor of digestion at lower temperatures. Maximum wet digestion temperatures are governed by the boiling point of the solution in which this digesting occurs, provided that the samples are not taken to dryness, and are roughly 400 to 500°C lower than the usual minimum dry-ashing temperatures.

The Kjeldahl procedure, with modifications, utilizes concentrated sulfuric acid as an oxidizing agent and has been by far the most widely used method for digesting plant and animal tissues. In addition, potassium sulfate or persulfate together with a catalyst such as copper, mercury, or selenium are generally employed. Perchloric acid may be used to great advantage with sulfuric acid as a modification of the original Kjeldahl method.

Nitric acid and mixtures of nitric acid with hydrochloric acid, hydrogen

peroxide, or perchloric acid have been successfully applied in wet-ashing procedures and are to be preferred whenever the introduction of excess sulfuric acid or sulfates is disadvantageous.

Extensive work has been carried out by Smith [5] and by Kahane [6] on the use of perchloric acid for the digestion of organic material. The former author's publication includes a review of the literature through 1940 and a compilation of some of the more important procedures employing this reagent in conjunction with sulfuric and with nitric acid.

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## CHAPTER 19

### THE SAFE HANDLING OF RADIOACTIVE MATERIALS

**19.1. Introduction.** The manipulation of radioactive materials presents two major problems with which a laboratory must be adequately prepared to cope in so far as they affect laboratory design, operative procedures, and personnel. These are, in the broadest terms, the health hazard of radiations to which personnel may be exposed and contamination of the laboratory and its equipment. Contamination represents a relatively uncontrolled source of radiation and in this sense must always be regarded as a potential health hazard in addition to the constant danger it poses for confusing experimental results. It demands, almost without exception when active materials are handled in appreciable quantities, that measures be taken to detect and eliminate it. So far as radiation is concerned, little differentiation can be made in the procedures taken to preclude any possibility of excessive personnel exposure, whether the radiation is from contamination or from carefully handled quantities of active material.

Fortunately, in most private institutions using radioactive materials in research and for therapy these problems are less complex than in the isotope processing laboratories of the Atomic Energy Commission because the quantities of active material handled are comparatively small. Laboratory design, equipment, and monitoring procedures for this reason often can be made quite simple. Usually the quantity of material will not exceed 100 millicuries, and rarely is it as great as a curie. Hot cyclotron targets in most instances are likely to be the most active sources to be dealt with and may at times approach the curie level. Almost without exception, however, when active material is handled in amounts greater than tracer quantities (~ microcuries) suitable protective measures must be instituted.

In addition to the quantity, the species of isotopes that will be handled in a laboratory affects the extent to which precautions must be taken. Isotopes with half-lives of a few hours or days do not present a problem of permanent contamination but usually represent serious health hazards by virtue of the high energy radiation associated with short half-lives. Materials with long half-lives are always serious contaminants in that they may seriously interfere with subsequent experimental results and may or may not be health hazards too. In particular, there is a group of isotopes which constitute a special health hazard irrespective of the energy or type of their radiation. This includes all isotopes with long half-lives that have a tendency



to remain fixed in the human body when breathed in the form of dust, ingested, or introduced through lesions. Among these, certain alpha emitters are the most dangerous because of the great damage inflicted on tissues by the heavy ionization produced by alpha particles.

The elaborate protective measures developed and practiced in the national laboratories where large quantities of radioactive materials are processed clearly demonstrate by their practically perfect record in health physics that every precaution taken to minimize the health hazard and danger from contamination is fully warranted. The general principles and much of the detail of these practices is now sufficiently well known to enable any laboratory contemplating the use of radioactive materials to institute the necessary procedures and install the proper facilities for its particular needs. The following sections are intended to outline these problems and practices, but reference to the literature cited is suggested for more exhaustive discussions.

**19.2. Medical Considerations.**<sup>1</sup> Perhaps the most obvious hazards to the health of laboratory personnel from radioactive materials are external irradiation by gamma rays and beta particles and internal irradiation from active material inadvertently taken into the body by inhalation, ingestion, or other means. The types of injuries or subsequent pathological changes following such exposures depend upon the radiation dose, the tissues irradiated, and various other factors such as the physical characteristics of the radiation involved and duration of exposure. The medical treatment of radiation damage is not well developed; some of the changes induced are insidious in onset, and some are associated with grave consequences. Since every effort must be made to ensure against significant exposure, emphasis should be placed on prophylaxis. The situation may be compared in some ways to the handling of infectious diseases in the laboratory or the use of poisonous reagents. Provided that guesswork as to radiation exposure is supplanted by measurement of radiation intensity and personnel monitoring and such possibilities as air contamination are explored, with adequate precautions taken, work may proceed with confidence and safety.

In addition to safeguarding laboratory personnel, thought should be given to the possibility of contaminating other areas and involving persons outside the laboratory, *e.g.*, by sending contaminated laboratory coats to the laundry or spreading activity through contaminated clothing and shoes. These considerations, the problem of radioactive waste disposal, and others are treated in the succeeding sections.

In many instances it is not possible to avoid some degree of external irradiation while working with radioisotopes; hence the need for knowledge of "safe" exposures or tolerance doses has arisen. Much has been written about this question, and reference may be made to a recent review by

<sup>1</sup> This section is by R. Lowry Dobson.

Parker [9]. By the tolerance dose is meant the amount of radiation that an individual may receive and suffer no observable ill effects. It is usually expressed as a dosage rate, as the amount allowable per day or per week. For several years in this country the accepted figure for x- and gamma rays has been 0.10 r per day. Factors taking into account the relative biological effectiveness of other types of radiation have modified this figure for neutrons, protons, and alpha particles. Table 40 gives the tolerance daily doses accepted by the Atomic Energy Commission for various types of radiation:

TABLE 40. DAILY TOLERANCE DOSES [10]

x-rays.....	0.10 r
Gamma rays.....	0.10 r
Beta particles.....	0.10 rep
Fast neutrons*.....	0.02 rep
Thermal neutrons.....	0.02-0.05 rep
Alpha particles.....	0.01 rep

\* Also applies to protons.

A lower tolerance figure has been advocated by some, and recently the National Advisory Committee on Radiation Protection has recommended 0.30 r per week for x- and gamma rays with proportionately lower figures for other types of radiation. The recommendation regarding exposure of the hands is that a dose three times that for total body irradiation be allowed, or approximately 1 r per week. This committee has also suggested that 300 r be taken as the upper limit for accumulated lifetime exposure. It seems probable that these lower allowable doses will be generally accepted in the near future.

The hazards associated with the presence of radioactive material inside the body depend upon the metabolism of the material, its radiation characteristics and half-life. Some substances, for example, when ingested, are not absorbed from the gut, others are readily absorbed; radium is a familiar example of a substance that may find its way to the blood stream by way of the gastrointestinal tract or the pulmonary tract and is deposited in bone. Its presence in the skeleton over long periods of time has been associated with the development of malignant bone tumors. Osteogenic sarcomas and anemias may be expected also with other long half-life isotopes that have affinity for the skeleton. Certain isotopes such as radio sodium are more widely distributed and result in general body radiation.

Care must be taken in the laboratory that radioactive vapors and dusts are not allowed to contaminate the air and result in internal radiation following inhalation. Obviously, smoking and eating in the laboratory are hazardous.

Fairly elaborate medical supervision of personnel has been carried out in Manhattan District and Atomic Energy Commission laboratories which

does not perhaps seem to be warranted in smaller laboratories where lesser amounts of activity are used. In the government laboratories periodic physical examinations, complete blood counts, and urinalyses have been done on all potentially exposed persons, as well as such special studies as examinations of the fingers for skin effects and assay of the urine for radioactivity as an indicator of internal deposition.

In laboratories where medicolegal considerations are taken into account, it would seem wise to provide for initial physical examination, chest x-ray, and clinical laboratory studies in an attempt to rule out the presence of malignancies or blood dyscrasias which might be linked to radiation exposure. Periodic examinations, especially of the blood, and terminal examinations would also seem indicated. In laboratories where these considerations are not important, it would nevertheless seem wise to provide, in addition to personnel monitoring with film badges or pocket electrosopes, at least for hematological observation of those workers who are potentially exposed to significant amounts of radiation or who may work with radiation for long periods of time.

There is no evidence that chronic exposure to radiation in the tolerance range will alter the normal blood picture, but blood counts may reveal abnormalities suggestive of radiation changes and lead to investigation of overexposures which might otherwise have gone unnoticed, or they may reveal incidental abnormalities such as anemia which should be corrected before work with radioactive material is undertaken.

In any case where there has been an accidental ingestion or inhalation of active material, or where it has been introduced into a wound, every effort should be made to prevent absorption or deposition of the radioactivity in the body. The methods employed will, of course, depend upon the circumstances, the type of material, etc. If the isotope has a long half-life and follows a metabolic pathway that keeps it in the body for long periods of time, the situation is very urgent. Copious washing of a contaminated wound is indicated immediately with consideration given to avoiding the spread of contamination to uninvolved areas; in rare instances, debridement may be warranted. Gastric lavage in cases of ingestion is indicated. Efforts should be made to determine from the material being handled exactly how great the problem is. Subsequent medical treatment will depend on individual circumstances.

**19.3. Laboratory Design.** The special features required in the design of a laboratory intended for radiochemical processes depend chiefly on the maximum level of activity that can be anticipated in future operations. When the quantity of materials handled approaches the 100-millicurie level, special precautions must be taken with regard to the health hazard in providing effective shielding and ease of decontamination. Although shielding can

be somewhat simpler for all activities at the microcurie level, or even at the millicurie level for materials that emit only beta or alpha particles, the danger of contamination should always be regarded as serious, particularly where long-lived isotopes are involved. Radiochemistry and other processing therefore should be confined strictly to those rooms prepared for these processes so that the spread of contamination by possible exchange of instruments, waste, clothes, etc., can be rigidly controlled. It is also clear that entirely separate rooms placed as far as practicable from active materials must always be maintained for radioassay and for experimentation with prepared active materials.

The most important feature is an adequately shielded and readily decontaminated hood. Concrete is probably the most economical material for this kind of structure, and it enables easy fabrication. The thickness of all sides including top and bottom should be great enough to attenuate the intensity of all gamma radiation to considerably less than that which will deliver a tolerance dose of 0.1 r per day at any accessible outside surface. The floor of the hood should slope downward to localize the spread of contamination caused by spillage, and it should be sufficiently large to enable nearly all operations with active materials to be performed under the hood. If the exhaust chimney passes through other portions of the building, particular care must be taken to ensure against leakage of fumes due to corrosion and also excessive accumulation of possible active condensates. Normally when volatile or gaseous active materials are processed in the hood, some means is provided for condensing and collecting the material.

In addition to a hood, each radiochemistry laboratory should contain a permanent vault or cove for storing all radioactive materials not in immediate use. Concrete or lead structures are the most suitable for this purpose. With properly designed compartments, each with its separate closure, access to one compartment and its contents is possible without danger of excessive radiation exposure from other materials stored in the vault. Wall thickness should be sufficient to attenuate gamma radiation to negligible intensity in all directions, and closures should be carefully checked with Geiger counters to detect possible leakage of radiation scattered through cracks or poorly fitting closures when high-intensity gamma emitters are stored.

Improvised storage coves hastily assembled with lead bricks should be avoided except in emergency since there is often a tendency to neglect bottom and back shielding and to overlook radiation leakage.

Additional permanent or semipermanent laboratory fixtures may include large general shields of lead surrounding areas or apparatus handling high-intensity gamma- and beta-active material.

Surface finishing materials for table and bench tops and other working areas may be expendable absorbing materials such as masonite or they may be

permanent, inert materials such as stainless steel or monel metal. Both types of surface material have certain advantages that must be considered according to the specific requirements of the laboratory when a choice is made. Absorbing materials, by taking up spilled or splashed liquid, do not allow radioactive dust to be readily formed. It can also be replaced at reasonable cost and effort after contamination. Stainless steel can be thoroughly cleaned, if necessary, with dilute acid, but it still has a tendency to allow dust to form unless the most diligent efforts are taken to avoid it. If contamination becomes serious, stainless steel is also expensive to replace.

All surfaces, particularly floors, should be nonabsorbing, smooth, and as free from cracks as possible. Wood floors for example are undesirable since it is impossible to avoid cracks between boards in their construction, and it is virtually impossible to remove subsequent contamination without replacing the floor. Concrete is also undesirable since it is impossible to decontaminate without removal.

**19.4. Special Laboratory Equipment.** Special laboratory equipment as well as procedures for handling radioactive materials must be designed primarily to safeguard personnel from irradiation while manipulating any pieces of apparatus containing active material. The variety and complexity of handling equipment that should be made available will be found to differ considerably from one laboratory to another and from time to time since the amount of protection from radiation which must be afforded by such equipment will depend upon the level of activity, the kind of radiation and, to some extent, the specific requirements of the handling procedure. Thus for high-intensity high-energy gamma-active materials, general shields enclosing the entire operation and remote handling devices are required, whereas for some isotopes that emit only soft beta particles, rubber gloves and shielding in air alone may suffice. For this reason the kind of handling equipment, shielding, and details of a process, however simple, should be carefully planned in advance. It is obviously pointless, expensive, and inefficient to employ great excesses of shielding and unnecessarily complicated handling equipment and procedures when a much simpler and more direct technique would provide the requisite protection. On the other hand the converse situation presents a positive danger when needless risks are taken to speed up work. Unfortunately there is as yet little standardization in the design of handling equipment, techniques, and apparatus for working with active materials, and consequently it has been the practice for each laboratory to prepare the instruments and shielding that its particular isotopes and processes require. Whatever the activity level and kind of radiation, however, the safest practice is that which always limits personnel exposure to little more than the normal background rate for cosmic radiations and other natural radiations.

Essential items of laboratory equipment are remote manipulating devices with which all pieces of apparatus, such as beakers, flasks, and serum bottles, either containing or near active material can be handled. Such devices serve two important functions. (1) They increase the distance of personnel from active material and thereby reduce the risk of exposure by allowing shields to be interposed, increasing the air absorption, and decreasing the intercepted radiation intensity by the geometrical factor of roughly the reciprocal of the distance squared. (2) Even the simplest of these instruments keep the hands and head away from regions above open vessels where direct and scattered radiation intensity is always the greatest.

The simplest devices for this purpose are scissors-type tongs, as shown in Fig. 119. With an assortment of lengths and clamp sizes they can be used for many general purposes, from manipulating stirring rods to screwing caps on bottles. Lengths up to 18 in. are the most useful; beyond this they become unwieldy.

Another popular form of tong is that shown in Fig. 120. The clamp is actuated by a drawrod or cable running through a long light metal tube to which the clamp is hinged. With the clamp closed by locking the drawrod under tension at the far end, both hands are freed to manipulate a piece of apparatus without excessive strain or caution. Devices of this kind can be made in almost any length and often can be manipulated with greater ease and assurance than scissors-type tongs. Convenient forms can be made for a wide variety of purposes such as turning stopcocks, handling glassware, and performing simple operations.

To these may be added many special devices as suggested by the mechanical requirements of a process. If a procedure is to be carried out routinely, it is often expedient to design special equipment to perform as much of the process as possible automatically and by remote control behind fixed shields. In such cases the efficiency of an operation is greatly increased, and the danger of exposure and contamination is reduced to a minimum. Examples of such processes and equipment are to be found in references 1 and 2.

Complex mechanisms such as the pantograph manipulator will doubtless be put to increasing general use as improvements in design make them more efficient and versatile and less unwieldy. To be useful for more than a few specialized operations they should permit several degrees of rotation and translation of the working end to be controlled easily from the remote operating end.

High-energy gamma-active materials in quantities greater than the 100-millicurie level necessitate handling behind general shields of sufficient thickness and area to stop all direct as well as scattered radiation. This requires the use of remote manipulating devices operating over the top or through the shield. These may take the simple form of strings actuating various pieces

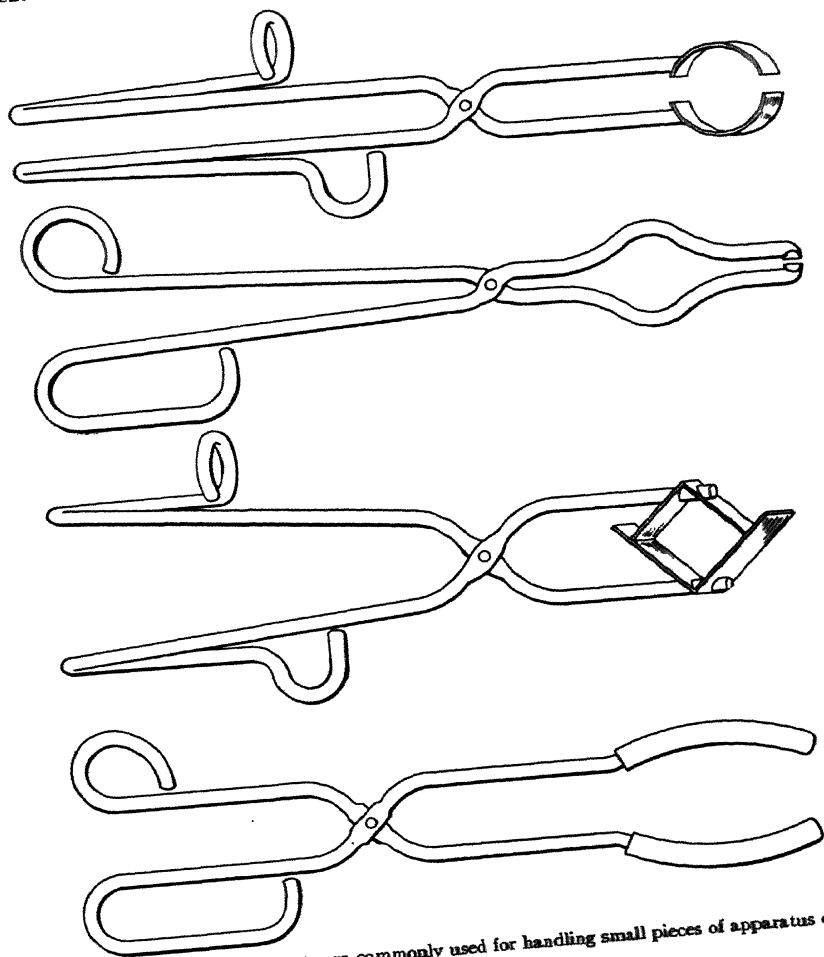


FIG. 119. Scissors-type tongs commonly used for handling small pieces of apparatus containing radioactive materials.

of apparatus, long tongs and rods bent to pass over the shielding, or they may consist in more elaborate mechanisms in the form of pantographs. Operations under these conditions usually must be performed with the aid of large mirrors in order to view the apparatus and manipulations indirectly through the shield. Two or four reflections are preferable since the mirrors easily

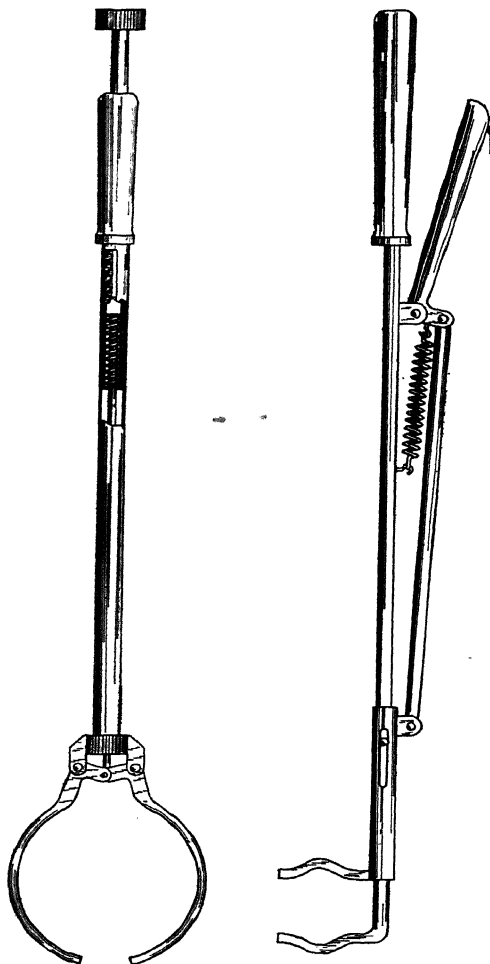


FIG. 120. Mechanical tongs for remote manipulation of radioactive materials.

can be arranged to give an erect image and a view of the apparatus the least confusing to the operator.

Laboratory coats and gloves are essential items for all personnel in a radiochemistry laboratory. Coats should never be worn outside the laboratory or be removed from it except for laundering, and if contaminated they must be destroyed. When high-activity materials or active liquids of any



kind are handled, the use of gloves is always compulsory. Thin rubber surgeon's gloves are to be preferred since they allow nearly unobstructed use of the fingers and can be thoroughly washed before they are removed or, if necessary, discarded. They are not intended to offer radiation protection but rather to protect the hands from direct contact with active materials and contaminated apparatus. Fabric and leather gloves should never be worn in the laboratory for they retain finely divided materials and absorb liquids. At best they are a greater hazard than bare hands. Similarly, lead-filled or shielded gloves are useless for most purposes because they are unwieldy to work with and secondary radiation produced in the lead may at times have a greater intensity than the primary radiation. This is especially true for very high-energy gamma rays. In certain instances, however, they offer the only means of protection as in cleaning or repairing heavily contaminated hoods, plumbing, etc., and then they are preferable to no protection at all.

Finally, the laboratory should be provided with suitable means of disposal of active waste. All contaminated paper, discarded apparatus, experimental animals, etc., should be placed in active waste cans provided only for this purpose. These are usually large standard trash containers made of corrosive-resistant metal, with tight-fitting covers conspicuously labeled with red paint. Since active waste liquids should never be washed down sink drains, they are best disposed of in glass or earthenware jars, shielded if necessary, and removed periodically or when the activity level at the outside surface of the container becomes too high.

**19.5. Safety Procedures.** Radiochemistry and all other operations involving appreciable amounts of active material should be carried out according to a definite set of safety rules covering procedures and personnel. Complete assurance that personnel will receive exposures no greater than the daily tolerance dose can be secured only when the safety rules established for a laboratory are strictly adhered to. Usually this can be accomplished (1) by thorough instruction of personnel in the dangers involved and the specific practices to be followed and (2) by rigid enforcement of the rules through periodic inspection and by posting a printed set of rules in a conspicuous place. When high activities are handled and an individual shows a persistent tendency toward carelessness and indifference, a job transfer normally is indicated before he or his associates are accidentally and seriously exposed.

The following rules, applicable to laboratories handling activities up to the curie level, are based largely on the rules and procedures practiced in the Clinton Laboratories of the Atomic Energy Commission. Taken as a complete set, the rules may be considerably more severe than is warranted for some laboratories and may be modified to suit less stringent radiation

conditions. Those rules which appear advisable regardless of the level of activity are indicated by asterisks.

\*1. All operations with active gaseous and hot liquid materials must be performed in well-ventilated hoods. When possible the entire process should be carried out in the hood and benches reserved for preparing reagents and apparatus.

2. Active material is never used in a procedure until a dummy run with inactive material has been made first and the procedure shown to be completely satisfactory. If changes are made in apparatus or the process, dummy runs should be repeated.

\*3. No procedure is permitted which involves danger of oral contamination whether by dust, liquid, or volatile materials. This is especially important for alpha activities and materials that remain fixed in the body.

4. Bench tops and hood floors are to be covered with absorbent paper before work is begun. Contaminated paper is to be removed and placed in active waste cans. In any case the paper should be replaced at regular intervals.

5. All glass receptacles containing highly active material must be placed and handled in stainless-steel trays, unless the receptacles have been provided with lucite shield containers.

6. Hot liquids must not be poured unless proper precautions are taken to contain spilled and spattered material.

\*7. All active materials not in immediate use are to be kept in the storage vault.

\*8. All covers and shields must be kept in place except when transferring materials.

\*9. Distillates must be condensed and collected if any active material is volatile.

10. All active materials entering or leaving the laboratory are to be monitored and pertinent data recorded.

\*11. Active material is never poured down sink drains but is placed in shielded jars to be properly disposed of.

\*12. Spilled or spattered material must be cleaned up immediately. Dust must be avoided by using wet cleaning materials which are then wrapped in clean paper and disposed of in the active waste cans. The area is to be monitored immediately thereafter.

\*13. Working areas are to be monitored before and after completion of work.

14. The laboratory is to be given periodic cleaning by a wet procedure.

15. Laboratory coats or coveralls must be worn in the laboratory but must be removed when leaving. Such apparel must be immediately replaced when contaminated.

\*16. Rubber gloves must be worn when directly handling receptacles containing active solutions.

\*17. Masks must be worn when working with gaseous or volatile active materials.

\*18. Pipetting solutions by mouth is strictly forbidden.

\*19. Smoking, eating, storing, or preparing food in the laboratory is forbidden.

\*20. Hands must be washed with soap and water before smoking and eating.

**19.6. Radiation Shields.** Shields and barriers intended for high-energy neutrons and gamma and beta radiation should be designed with three factors in mind: attenuation of direct radiation; emission of secondary radiation, including both x-rays and electrons; and scattered radiation. Adequate thickness of shielding material eliminates the first two factors. Insufficient thickness, it should be noted, can be worse under certain conditions than no shielding at all since the intensity of secondary radiation produced in the shield may be many times greater than the primary and its secondary radiation in air alone. The extreme case in point is encountered when the shield thickness just equals the transition depth for the gamma rays or fast neutrons. Scattered radiation is sometimes difficult to predict but can be eliminated by adequate coverage and by avoiding cracks and poor joints in shields. In all cases when shields are assembled, the shield surface and working area should be monitored before personnel are permitted to work.

Concrete, because it is comparatively inexpensive and structures are easily fabricated, is the most useful material for large shield structures such as hoods and storage vaults. But because of the large variety of shields needed in diverse procedures, lead is more convenient for most other shielding purposes. It is usually made available in some convenient standard sizes of plates and blocks small enough to be handled easily. For example, the lead shield stock may include plates measuring 12 by 6 by  $\frac{1}{2}$  in. and blocks 3 in. square or 6 by 3 by 2 in. With these bricks and plates, shields of any size and shape can be quickly erected for a specific operation.

Estimates of shield thickness are easily obtained when the maximum energy of the radiation and the absorption coefficient or half-value thickness are known. If  $I_0$  is the intensity or dosage rate at the inside surface of the shield, the intensity  $I$  of the primary radiation at the outside surface is

$$I = I_0 e^{-\mu x}$$

where

$\mu$  = absorption coefficient

$x$  = thickness

or in terms of half-value thickness  $T$  the transmitted intensity through  $n$  half-value layers is

$$I = I_0 2^{-n}$$

Conversely when it is known that the dosage rate or intensity must be reduced by a factor  $f$ , the number of half-value layers required for a specified absorbing material and energy is

$$n = 3.3 \log f$$

These formulas, in a strict sense, apply only to collimated beams of radiation and hence will give somewhat conservative values for most conditions met in practice. The requisite reduction could be achieved by distance alone, for at distances large compared to the largest dimensions of the volume of active material, the intensity diminishes as the distance squared. Thus if  $I_0$  is the dose rate or intensity at distance  $d$ , the intensity at any other radial distance  $x$ , neglecting absorption, is

$$I = I_0 \left( \frac{d}{x} \right)^2$$

Materials that emit beta or alpha particles but no gamma radiation frequently can be handled with shielding confined to each piece of apparatus. This is especially useful and effective in the form of separate thick-walled lucite containers tailored to fit constantly used items of glassware such as beakers, flasks, serum bottles, and test tubes [1], thus enabling the operator to see and handle pieces of equipment directly. To be useful, however, such containers must be as completely closed as possible, preferably with screw-type caps or plugs. Transfer of active liquids is made through a small hole in the cap or by removing the cap temporarily. At these times no part of the body, including the hands, should be above and near the level of the opening since the daily tolerance dose can sometimes be received in a matter of seconds from the emerging cone of intense direct and scattered radiation. The container design should also be such that contamination of the outside from spilling or spattering is minimized. Before reuse they should be thoroughly cleaned with dilute acid. Aside from this type of shield, lucite and other transparent plastics offer limitless possibilities for inexpensive and efficient protection against beta-active and even low-intensity, soft gamma-active materials. The many uses to which they can be put to simplify operations while still providing the necessary protection will depend on the ingenuity exercised in designing special shields.

Boron, cadmium, and bismuth are the most effective materials for shielding against slow neutrons. They have large resonance and thermal neutron capture cross sections (boron,  $\sigma \sim 500$  barns; cadmium,  $\sigma \sim 7,000$  barns; bismuth,  $\sigma \sim 10$  barns) and can be obtained in quantities and forms suitable for fabricating shields. Boron and cadmium should in addition be backed with gamma-ray absorber. Bismuth alone does not emit gamma rays in the process of neutron capture. Fast neutrons present a shielding problem

similar to that attending gamma radiation in that large thicknesses are necessary and scattered and secondary radiation can be serious. The neutrons first must be slowed to near the thermal velocities and then absorbed in a thin layer of boron, cadmium, or bismuth. Shielding materials for fast neutrons therefore should be low in atomic weight. Large barriers constructed of concrete or water are customarily employed because they are the least expensive and easy to fabricate. For smaller shields, for example, to shield Ra-Be sources, paraffin or graphite is more convenient.

TABLE 41. GAMMA-RAY ABSORPTION COEFFICIENTS OF SHIELD MATERIALS [6]

$E_\gamma$ , mev	Absorption coefficient, $\text{cm}^{-1}$			
	H <sub>2</sub> O	Al	Fe	Pb
0.2	0.14	0.33	1.06	5.0
0.5	0.090	0.23	0.63	1.7
1.0	0.067	0.16	0.44	0.77
1.5	0.057	0.14	0.40	0.57
2.0	0.048	0.12	0.33	0.51
2.5	0.042	0.10	0.31	0.48
3.0	0.038	0.090	0.30	0.47
4.0	0.033	0.082	0.27	0.48
5.0	0.030	0.074	0.24	0.48

TABLE 42. HALF-VALUE LAYERS OF SHIELDING MATERIALS FOR GAMMA RAYS [6]

$E_\gamma$ , mev	Half-value layer, cm			
	H <sub>2</sub> O	Al or concrete*	Fe	Pb
0.2	5.1	2.1	0.66	0.138
0.5	7.8	3.0	1.11	0.42
1.0	10.2	4.5	1.56	0.9
1.5	12.0	5.1	1.74	1.2
2.0	14.4	5.7	2.10	1.35
2.5	16.5	6.9	2.22	1.47
3.0	18.3	7.8	2.31	1.47
4.0	21.0	8.4	2.55	1.47
5.0	23.1	9.9	2.88	1.47

\* Poured concrete with limestone aggregate, density 2.7.

**19.7. Radiation Monitoring.** Rigid control and assessment of radiation exposure can be ensured only through diligent surveys of radiation intensity in all areas and equipment to which personnel have access and through the continuous indication of each individual's total exposure. In view of the

many possibilities that exist for otherwise undetected contamination and radiation leakage, no preparations and procedures, however cautiously planned to guard against exposure, can be considered safe until every possible type and source of radiation has been measured. This requires also that careful monitoring be practiced as a part of normal laboratory operations. Because of the insidious nature of radiation in that no immediate effects are apparent and the fact that the time and effort spent in monitoring do not contribute to the work at hand, there is sometimes a tendency toward leniency or even disregard for adequate monitoring. This fact alone makes it imperative to enforce strictly the monitoring regulations that are adopted by a laboratory.

The principal techniques and instruments used at the present time for monitoring activities up to the 100-millicurie level are described below. For the most part they are adapted from the methods employed in the laboratories of the Atomic Energy Commission. Although they are essential to all levels of activity and adequate for levels of activity handled in most private institutions, at very high levels, additional and more elaborate measures must be taken and reference to the declassified project literature listed at the end of this chapter is suggested.

*a. Film-badge Meter.* Film badges have been widely adopted as personnel exposure indicators wherever radiations are encountered. They should be worn constantly by all personnel, including visitors, in areas likely to contain contamination, active material, or radiation fields from high-energy accelerators. When a permanent record of each person's film exposure is kept, it provides a necessary history of his total exposure as well as legal protection for the institution in suits over injuries claimed to have resulted from excessive radiation exposure.

Beta-particle and gamma-ray film meters usually consist of dental film mounted in a metal holder designed to be clipped on outer clothing such as the lapel or sleeve. Two films are sometimes inserted, one sensitive in the dose range from 20 to 3,000 milliroentgens and the other in the range 0.5 to 20 r [3]. The response of the emulsions is nearly proportional to the roentgen dose and is practically independent of gamma-ray energy over a wide range. There is, however, usually a marked increase in sensitivity for very soft gamma rays in the energy range 0.05 to 0.3 mev where the sensitivity is greater by a factor of  $\sim 20$ . Part of the film is covered by a cadmium mask thin enough to pass gamma rays with energies greater than  $\sim 75$  kev, and the remainder of the film is left uncovered except for the paper wrapping to detect beta particles. The darkening of the uncovered portion, however, is not always a reliable measure of beta exposure if soft gamma-ray exposure has been received as well. Also darkening behind the cadmium shield can be produced by the reaction  $\text{Cd}(n, \gamma)\text{Cd}$  on exposure to slow neutrons. After

development the indicated accumulated dose is determined by densitometer measurement of the film darkening.

Neutron film meters are similar to gamma-ray meters except in the use of proton-sensitive emulsions. The area behind the cadmium registers only recoil proton tracks due to fast neutrons, while the uncovered area registers both proton recoils and protons from the reaction  $N^{14}(n, p)C^{14}$ . Dose is estimated from dark-field microscopic examination of the number of tracks per unit area. The weekly tolerance dose of fast and slow neutrons is approximately 4,500 tracks per square centimeter [3,5].

Film meters have been used in other forms such as rings worn on a finger. These and other forms are particularly useful in working with beta emitters when there is danger of excessive hand dose which would not be registered on more remote film meters.

*b. Pocket Meters.* Two forms of pocket meters have been developed for registering personnel exposure to gamma radiation. The first of these is a pen-sized air ionization chamber, or air condenser, charged to approximately 150 volts before it is worn. It is read, preferably at the end of each day, by discharging it through a minometer or a string electrometer. Since it is calibrated for only one gamma energy, a considerable error can result from exposure to gamma radiation of very different energy. The useful range is approximately 0 to  $\frac{1}{4}$  r, and saturation can occur at high radiation levels. These meters are subject to accidental discharge from rough treatment, high humidity, and insulator leakage and should accordingly be treated with reasonable care.

The second meter is a pen-sized dosimeter consisting of a fiber electrometer with a scale and magnifying lens at one end with which to view the fiber position. Once charged, the dose received can be read at any time. These instruments are useful only for high radiation intensities and are subject to the same disadvantages as the air condenser with regard to accidental discharge and false dosage indication.

*c. Air Contamination.* Surveys of air contamination due to radioactive dust are especially important in areas handling appreciable quantities of alpha emitters or those particularly hazardous beta-gamma emitters which when ingested or breathed remain fixed in the body. The maximum permissible level for each isotope is somewhat uncertain, but for plutonium the Clinton Laboratories have set the limit at  $3 \times 10^{-11}$  microcurie per cc, and for beta emitters, the value  $\sim 10^{-7}$  microcurie per cc.

Both precipitators and air filters are used for air monitoring. The former consists of a cylindrical aluminum foil in which dust is precipitated electrostatically as air is drawn through at a known uniform rate. From Geiger-counter measurements of the accumulated alpha, beta, and gamma activity on the unrolled foil, the air contamination can be estimated. The second

instrument measures contamination by drawing air through filter paper surrounding a thin-walled Geiger counter. The counter output can be applied to a recording instrument which then gives a continuous record of accumulated activity. The most useful correlation between accumulated activity and dose received by the lungs is had when the rate of air flow in either instrument above is made equal to the average lung minute volume of approximately 8 liters per sec.

*d. Radiation Fields.* Appreciable intensities of gamma radiation and fast neutrons over large areas are sometimes encountered when, due to inadequate shield thickness, direct radiation is not sufficiently attenuated or when the shielding, improperly constructed, allows scattered radiation to leak out. Radiation fields such as these are quickly and easily detected when radiation surveys are diligently carried out. This is best accomplished through routine monitoring while work with active materials is in progress and by careful surveys of new shield assemblies before work is started. Only in this way can it be made certain that shielding is sufficient and does not scatter or transmit radiation into easily accessible areas. In no instance should shielding, including the storage vault and hood, be regarded as safe for a large quantity of active material until a survey has been taken.

Portable ionization chambers and electroscopes, now available commercially in many forms, are probably the most dependable devices for gamma-ray detection in open areas. However, the greater sensitivity and faster response of portable counter tubes can often be used to advantage in detecting radiation leakage through cracks and joints in shielding. Fast neutrons are detected most effectively with hydrogen-recoil ionization chambers and counters, while slow neutrons are best detected with boron trifluoride-filled or boron cathode counters.

Beta radiation as such cannot be considered as a possible hazard at considerable distances from active material, but in the immediate vicinity of apparatus containing the active material the intensity can be great enough to deliver the daily tolerance dose in a few seconds exposure. When it is necessary for the hands to have access to apparatus, particularly above open containers, it is extremely important to measure the dosage rate before that part of the procedure is undertaken. The most convenient monitoring instruments for this purpose are portable thin-window counters; either cylindrical-tube or bell-type counters may be used effectively. These instruments should be conveniently at hand in the laboratory at all times, and especially is this important when new procedures are being worked out or when particularly active quantities of material are handled.

*e. Surface Contamination.* Contamination of working surfaces is inevitable at one time or another, and in hoods it is practically unavoidable. Even when the surface activity delivers less than the daily tolerance dose, the



contamination should be regarded as a hazard particularly if it is alpha active since it becomes a potential source of radioactive dust which then may be breathed or ingested. More often the principal objects of contamination are equipment and apparatus used in handling and in radiochemistry. Shoes, hands, and laboratory coats are also subject to contamination and should be monitored when contamination is possible.

Surface monitoring normally is the most important function of routine surveys and should in part also be made a responsibility of the persons using apparatus and work space. Periodic surveys should include measurements of the alpha, beta, and gamma activity of all permanent surfaces such as floors, workbenches, hoods, telephones, sinks, etc., where active material

TABLE 43. RADIOACTIVE-CONTAMINATION TOLERANCE LEVELS\*

Object	Tolerance Level
Hand.....	1 mr/hr of $\beta$ and $\gamma$
Shoe: outside.....	14 mr/hr of $\beta$ and $\gamma$
inside.....	$\frac{1}{2}$ mr/hr of $\beta$ and $\gamma$
Clothing.....	$\frac{1}{2}$ mr/hr of $\beta$ and $\gamma$
	1,500 disintegrations/min of $\alpha$ from area of 150 cm <sup>2</sup>
Thyroid.....	1,000 mr/24 h in thyroid
Table tops, floors, etc.....	0.1 mr/hr with counter at surface
	2,000 disintegrations/min of $\alpha$ from area of 150 cm <sup>2</sup>
Smear tests on any surface:	
2 sq. in. filter paper smeared	200 cpm of $\beta$ and $\gamma$ ;
over 12 sq. in. and counted	0 disintegrations/min of $\alpha$
with $\alpha$ , $\beta$ , and $\gamma$ counters	

\* These tolerance levels are based on values established for the Clinton Laboratories of the Atomic Energy Commission, Oak Ridge, Tenn., and reported in references 1, 3, 4, and 7.

could be spread. When a surface shows an activity higher than the tolerance level, it should be posted with signs and necessary remedial steps taken as soon as possible.

Part of the responsibility for monitoring working surfaces and equipment is placed with the person working with them. The suggested procedure as practiced in some laboratories requires that such persons survey the work surfaces and apparatus before work is begun and again when work is completed, or at the end of his shift. This also applies to glassware and other equipment that has just been cleaned. After a spill, in hoods as well as on benches and floors, the affected area should be carefully surveyed after it has been cleaned, and it should not be considered decontaminated until a survey shows that the activity is no greater than the tolerance level.

A great variety of portable survey instruments have been developed which are useful for surface-activity measurements, and some of these are now commercially available. They include electroscopes with thin windows suitable

for beta and gamma radiation, air ionization chambers, and counters. Air ionization chambers are particularly useful for detecting alpha particles and very soft beta particles since there is no intervening absorber between the surface to be measured and the sensitive volume of the chamber. Geiger counters with thin windows may serve the same purpose and are more sensitive in some cases, but generally they are more useful for energetic beta particles and gamma rays. Either the bell or cylindrical types can be mounted in appropriate holders to enable the operators to scan areas by moving the counter with the window immediately adjacent to the surface. Both ionization chambers and counter tubes are sometimes fitted with sliding filters which can be moved over the window. With no filter in place, alpha and beta particles are registered; with aluminum foil, only beta and gamma radiation are admitted; and with sufficient thickness of bakelite, lucite, or other plastic, alpha and most beta particles are stopped and only gamma radiation is registered.

**19.8. Shipping Regulations.** On the basis of investigations and suggestions submitted by the subcommittee on shipment of Radioactive Substances of the committee on Nuclear Sciences of the National Research Council,



FIG. 121. Label required for outside shipping container of Groups I and II radioactive materials. Red printing on white paper measuring 4 by 4 in. square.

the Interstate Commerce Commission adopted regulations covering the packaging, labeling, and transportation of radioactive substances within the United States. The regulations are designed to minimize the possibility of radiation exposure to personnel and undeveloped photographic emulsions while the active material is in transit or storage. Full details of the regulations are given in Supplement No. 10 to I.C.C. No. 4670, ME-I.C.C. No. 4076, or the *Federal Register*, Vol. 12, No. 220, pp. 7329-7333 [see also reference 8]. The principal packing and shielding regulations governing the shipment of those radioactive materials and the quantities normally required for experimental and therapeutic uses are outlined below.

Radioactive material, any material emitting ionizing radiation, is listed as Class D poison and divided into three groups:

Group I. "Radioactive materials that emit gamma rays only or both gamma and electrically charged corpuscular rays." Each outside container must be labeled with a properly executed red label, as shown in Fig. 121.

Group II. "Radioactive materials that emit neutrons and either or both the types of radiation characteristic of group I materials." Each outside container must be labeled with a properly executed red label, as shown in Fig. 121.

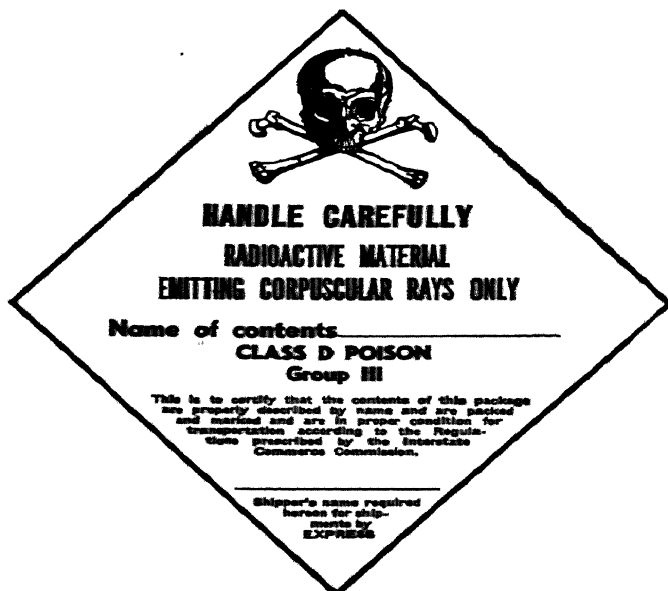


FIG. 122. Label required for outside shipping container of Group III radioactive materials. Blue printing on white paper measuring 4 by 4 in. square.

Group III. "Radioactive materials that emit electrically charged corpuscular rays only . . ." Each outside container must be labeled with a properly executed blue label, as shown in Fig. 122.

Summary of the principal shipping regulations is as follows:

*a.* Radioactive materials that have a tendency to remain fixed in the human body must be packed in an inside tube container of stainless steel, malleable iron, or brass with dimensions not to exceed 3 in. diameter or 8 in. length. It must have a minimum wall thickness of  $\frac{1}{8}$  in. and a screw-type cap or plug at one or both ends with a number of threads per inch not less than that of U.S. Standard taper pipe threads.

*b.* Packing and shielding must be such that film fogging in 24 hr at 15 ft. will not exceed that produced by 11.5 milliroentgens of gamma rays from radium shielded by  $\frac{1}{2}$  in. of lead.

*c.* There must be no significant surface contamination.

*d.* Minimum dimension of outside shipping container is 4 in.

*e.* Gamma radiation must not exceed 200 milliroentgens per hr at any readily accessible surface. Container must be equipped with handles or protective devices if necessary to meet this condition.

*f.* Outside container must be wood or fiberboard as per specifications or else as approved by the Bureau of Explosives.

*g.* Shielding of group I materials must be such that gamma radiation at 1 m from source never exceeds 10 milliroentgens per hr and corpuscular radiation does not escape to exterior of outside container. The shield must not open or break under conditions incident to shipping.

*h.* Group II materials must be packed in suitable inside container and shielded so that radiation at right angles to long axis does not exceed 10 milliroentgens per hr at 1 m for gamma radiation or the equivalent in corpuscular radiation or 2 milliroentgens per hr at 1 m gamma equivalent of neutrons. If several types of radiation are present, the total is not to exceed these values.

*i.* Liquid materials of all groups must be contained in tight glass, earthenware, or other suitable inside container and surrounded by sufficient absorbent material within the shield to absorb the entire liquid contents. Alternatively, the metal inside container of Rule *a* can be used.

*j.* Group III materials must be so shielded that primary corpuscular radiation does not reach the exterior of the shipping container and secondary radiation at container surface does not exceed 10 milliroentgens per 24 hr.

Radioactive materials are exempt from prescribed packing and labeling provided:

*a.* No leakage can occur under conditions incident to transportation.

*b.* The package must contain not more than 0.1 millicurie of radium or polonium, or that amount of  $\text{Sr}^{89}$ ,  $\text{Sr}^{90}$ , or  $\text{Ba}^{140}$  which disintegrates at the

rate of 5 million atoms per second, or any other material that disintegrates at the rate of 50 million atoms per sec.

c. There must be no significant corpuscular radiation at the package surface, and gamma radiation must not exceed 10 milliroentgens per 24 hr.

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## CHAPTER 20

### THE ELECTROSTATIC GENERATOR

**20.1. Description.** The most successful form of electrostatic generator for the acceleration of heavy charged particles is that first proposed by Van de Graaf [1] and subsequently developed by Van de Graaf and others [2-6]. High potentials are produced by the accumulation of charge on large closed-surface conductors, often spheres, called terminals. The terminals are charged to the desired electrostatic potential by means of an endless belt of nonconducting material serving as a charge conveyer. Charge is sprayed on at the grounded end of the belt and subsequently removed by a collecting comb in the terminal. The charge collected appears only at the surface of the terminal, leaving the interior a field-free region as required by elementary principles of electrostatics. Ions formed by a source within the terminal are accelerated through a vacuum tube running from the terminal to ground or to a second terminal charged to the opposite polarity. The maximum energy

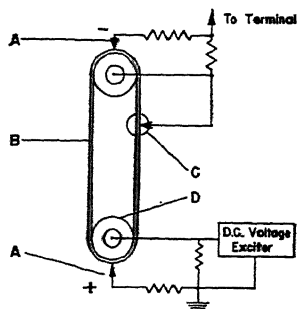


FIG. 123. Charging system for electrostatic (Van de Graaff) generator. A, spray combs; B, charging belt; C, collector and shield; D, metallic pulley.

imparted to a particle is  $eV$ , or in terms of electron volts  $nV$ , the charge number of the particle times the potential difference between the ends of the accelerating tube. This places a severe limit on the energies derivable from such machines as compared to resonance and induction accelerators since the maximum energy to which a particle can be accelerated is only equal to the voltage to which the terminals can be charged. The maximum voltage is established when the rate of charge leakage by corona discharge from the terminal equals the transport of charge by the belt. In the largest units thus far built this potential has exceeded 5 million volts [2].

**20.2. Construction and Operation.** A typical belt charging system is shown in Fig. 123. To achieve the highest equilibrium voltage requires the greatest practicable charge transport; belts are operated at saturation charge, made as wide as the mechanical design permits, and operated at the highest practicable speed—usually several thousand feet per minute. Cotton and paper belts have proved to be more practicable in large installations than silk, rayon, or linen, partly because of their consider-

ably lower cost but also because of better electrical properties [1.3]. Charge is sprayed on the belt by a spray wire stretched across the belt at a distance of approximately  $\frac{1}{2}$  in. and charged to a potential ranging from 5 to 20 kv as determined by the desired equilibrium voltage on the terminal. The charge conveyed to the terminal is then removed with an efficiency of 35 to 60 per cent by a collecting comb consisting of a metal strip containing numerous

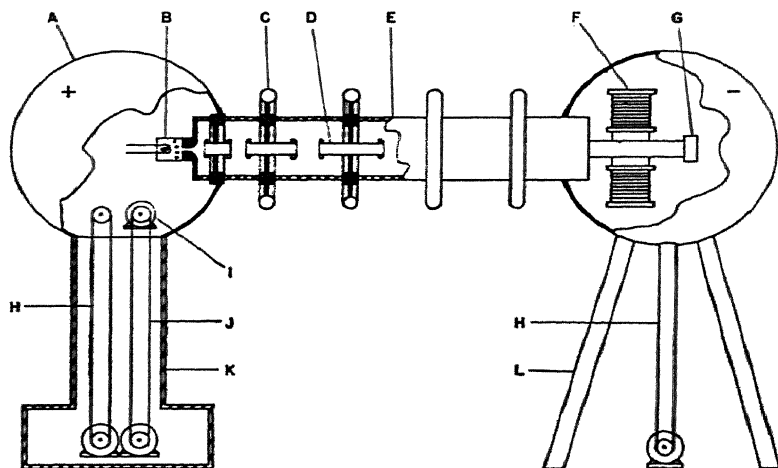


FIG. 124. Schematic diagram of the Van de Graaff electrostatic generator. *A*, terminal: aluminum shell; *B*, ion source: filament, grid, and capillary cathode through which ions pass into accelerating tube; *C*, corona ring; *D*, drift tube; *E*, accelerating tube insulator section; *F*, electromagnet for magnetic separation of ion beams; *G*, target; *H*, charge belt; *I*, power generator for source and other equipment; *J*, auxiliary belt for driving power generator; *K*, cylindrical type support insulator; *L*, tripod type support insulator.

sharp points directed toward the belt near the terminal pulley. Charge of opposite sign again is sprayed on the belt in the terminal and removed at the ground end, thus doubling the terminal charging rate. The current delivered to the belt by a charging system has been shown to be approximately [1]  $I = k(V - V_0)^2$ , where  $k$  is an empirical constant,  $V$  is the spray-wire voltage, and  $V_0$  is the spray-wire threshold. The threshold voltage for sharp-pointed combs is zero; for wires of small diameter, it is usually several kilovolts.

The vacuum accelerating tube, illustrated in Fig. 124, is constructed of a series of insulating sections made of glass, pyrex, or porcelain which are separated by metallic rings and vacuum-sealed. The metal rings support external corona rings, which ensure uniform potential distribution along the

length of the tube, and internal drift tubes through which the ion beam passes.

The drift tubes are essential for their focusing effect on the beam in order to compensate the defocusing caused by the initial angular divergence of ions leaving the ion source, space charge spreading, and scattering by residual gas in the accelerating tube. The focusing arises from the form of the electrostatic field in the gap between two successive drift tubes. As shown in Fig. 125, the bowed field in the first half of the gap tends to deflect all ions

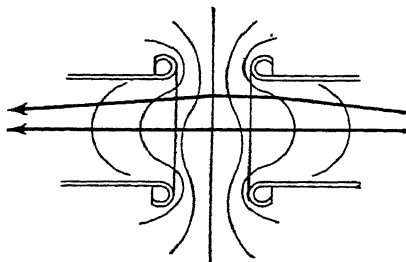


FIG. 125. Electrostatic focusing field between drift tubes (equipotential lines are shown). Trajectories of axial and off-axis particles shown by heavy lines.

toward the tube axis. Although the effect in the second half of the gap is reversed because of symmetry of the field, the ions have in the meantime gained axial momentum and the angle by which they are deflected from their path is then smaller than the initial focusing effect. The net effect, therefore, is to maintain a small pencil of ions that emerges with a small cross-sectional area at the target. It is apparent from the principles of the process that the crucial focusing takes place in the first few gaps because the

deflections are largest when particles are moving with comparatively low velocity.

The principal structural features of this type of accelerator are a high order of insulation, shielding, and a good geometrical form for the terminals to reduce heavy charge loss by excessive corona discharge and sparking. Most such generators have been constructed with terminals of nearly spherical form from aluminum sheet, spun or otherwise fabricated to the desired shape. Insulation is provided for by mounting the terminal on sufficiently long tubes of Herkolite, Textolite, or other nonconducting materials which can be fabricated in large structural form. Power for operating the ion source and auxiliary equipment in the terminal is derived from a generator mounted within the terminal and driven by either the charging belt or an auxiliary belt. When possible, humidity control is provided in the space occupied by the charging belt since a relative humidity of less than 50 per cent is generally necessary for consistent and optimum operation.

An important improvement in the design and operation of electrostatic generators was introduced by enclosing the entire unit in a pressure chamber filled with air, nitrogen, carbon dioxide or freon ( $\text{CCl}_2\text{F}_2$ ) at high pressure. The chief advantage lies in the greater insulator strength of gases at pressures above atmospheric, with a corresponding decrease in the break-



down gap for a particular voltage. This permits a very considerable reduction in the physical dimensions which become an important consideration in high-voltage installations. It allows, in addition, rigid control of the atmosphere and, in particular, the humidity. The essential features of a high-pressure electrostatic generator are the same as in units operated at atmospheric pressure. They are however built compactly and enclosed in a steel tank designed to withstand a pressure of 10 atm or more. Because of the small spacing between the terminal, accelerating tube, and the grounded tank wall, it is necessary to shield the entire unit with closely spaced hoops of aluminum tubing to ensure a uniform potential distribution and to reduce sparking to the tank wall.

The acceleration of protons and alpha particles in an accelerator of this type does not provide strictly homogeneous beams of particles. Hydrogen, for example, introduced into the ionization chamber is injected into the accelerator tube both as  $H^+$  and  $H_2^+$ . Both particles reach the target with the same kinetic energy, but the molecular ion splits on impact into two  $H^+$  particles, each with only half the original kinetic energy. Similarly, when accelerating alpha particles, both  $He^+$  and  $He^{++}$  are produced by the source. Energetically homogeneous beams can be obtained readily, however, by passing the accelerated beam through a magnetic field, or analyzer, to separate the particles  $H^+$  and  $H_2^+$  or  $He^+$  and  $He^{++}$  before they reach the target.

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## CHAPTER 21

### THE CYCLOTRON

**21.1. Description.** The cyclotron, or magnetic resonance accelerator, first proposed and developed by Lawrence [1], enables high energies to be imparted to heavy charged particles by repeated accelerations across a comparatively small electric field. Ions formed at the center of a uniform magnetic field of 10,000 gauss or more are accelerated in the gap between two hollow D-shaped electrodes, known as dees, to which a high-frequency potential of 10 to 100 kv is applied in opposite polarity, as shown in Fig. 126. An

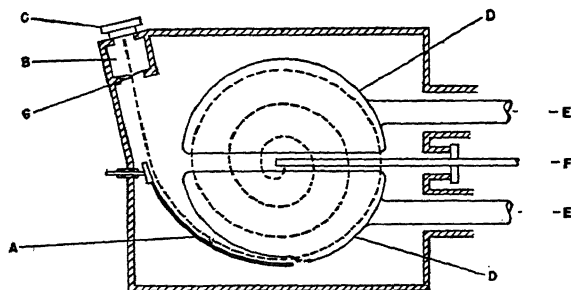


FIG. 126. Schematic diagram of the cyclotron. *A*, deflector; *B*, vacuum lock used without foil window for internal target; *C*, target; *D*, dees; *E*, dee stems; *F*, filament leads to ion source at center; *G*, aluminum foil used as a vacuum seal for external targets.

ion starting from the center of the magnetic field is constrained to move in nearly circular orbits of slowly increasing radius while accelerated by successive impulses in traversing the gap between the dees. Synchronization of the high-frequency electric field with the frequency of rotation of the particle ensures that the maximum dee voltage occurs at, or just before, the instant the particle passes the gap. During the remainder of its path in the field-free region within the dees, the particle is unaffected by the changing electric field which has reversed its polarity when the particle again enters the next gap. Two impulses, therefore, are experienced by a particle in each cycle.

The magnetic field does not contribute to the energy of an accelerated particle and serves only to constrain the particle to a circular path whose radius at any instant depends on the velocity.

$$r = \frac{mv}{eH} \approx \frac{1}{H} \sqrt{\frac{2m\pi V \sin \varphi}{e}}$$

where  $m$  = mass of particle

$v$  = linear velocity

$e$  = charge of particle

$H$  = magnetic field strength

$n$  = number of traversals of gap

$V$  = peak radio-frequency voltage

$\varphi$  = phase of particle: angular position from gap when  $V = 0$

The angular velocity of the accelerated particle is independent of energy and linear velocity, provided that the relativistic increase in mass is negligible. The period of rotation is then constant, and its magnitude is given by

$$T = \frac{2\pi mc}{eH}$$

The frequency of the particle, as well as that of the oscillating electric field, is, correspondingly, constant with a magnitude given by

$$\omega = \frac{1}{T} = \frac{eH}{2\pi mc}$$

The highest energy to which a particle can be accelerated is determined, aside from the limitation imposed by the relativistic increase in mass, by the magnetic field strength and the maximum orbital radius.

$$E_{\max} = \frac{e^2 R^2 H^2}{2mc^2}$$

where  $R$  = maximum radius

$c$  = velocity of light

If the frequency and peak voltage applied to the dee are kept constant and only the magnetic field is altered for the acceleration of particles of different mass, the maximum energy to which the particles can be accelerated for a given radius is directly proportional to the mass.

$$E_{\max} = 2\pi^2 m \omega^2 R^2$$

After the particles have been accelerated to the maximum energy and radius, extraction of the beam is accomplished, for energies up to 50 mev at least and possibly higher, by electrostatic deflection of the particles from their normal orbit. A long curved plate located a quarter or third of the distance along the periphery of the orbit is operated at a sufficiently high negative potential to deflect the beam out of its normal orbit and into the region where the magnetic field begins to fall off rapidly. In this way the beam can be directed into a tangentially located target chamber and if necessary brought out of the vacuum tank through an aluminum-foil window.

Alternative schemes have been proposed which may prove more successful in extracting very high-energy beams ( $> 100$  mev) where simple electrostatic deflection is impracticable. For instance, if the magnetic field is locally weakened by shims along a spiral path leading outward from the maximum radius, ions of the proper energy entering this region can be led out to the

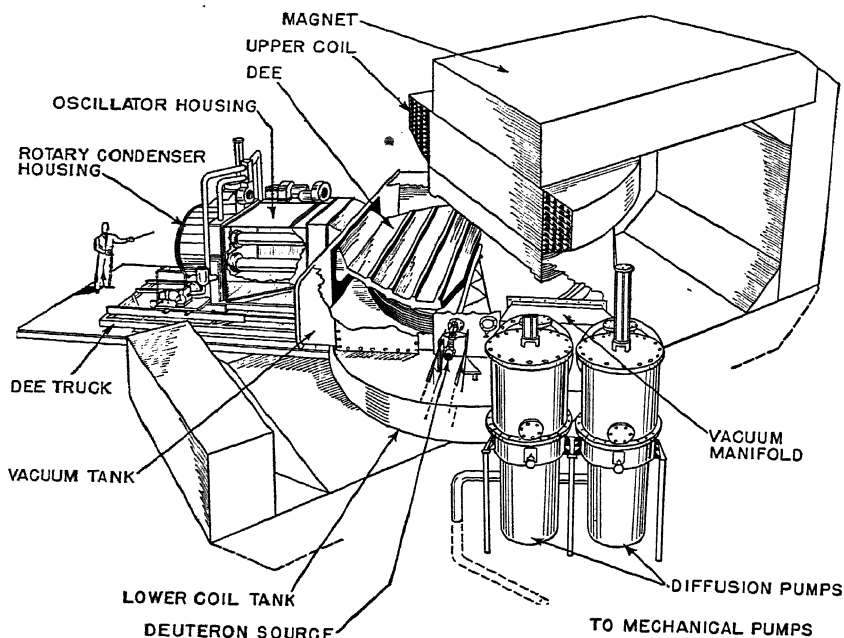


FIG. 127. Cut-away diagram showing the main structural features of the 184-in. frequency-modulated cyclotron of the Radiation Laboratory, University of California. [Reprinted by permission of the Radiation Laboratory.]

periphery of the field [2]. Magnetic deflection of this kind combined with appropriate electric fields may ultimately provide the most satisfactory deflector system in such cases.

The general structural features of the cyclotron are illustrated by the diagrams of the 184-in. cyclotron at the Radiation Laboratory of the University of California shown in Figs. 127 and 128.

**21.2. Ion Paths.** The ultimate fate of an ion depends largely on its trajectory during the first few cycles after it leaves the source. Less than  $10^{-4}$  of the ions produced at the source ever reach the target. The majority of ions are lost immediately by neutralization in collisions with parts of the

source and the dees, and partly by recombination. Furthermore, ions can be started on stable orbits during only a part of the electric-field cycle. Those which start long before the dee voltage has reached a maximum value are ultimately defocused and strike the dees; those starting long after the dee voltage has passed a maximum value may be accelerated and find stable orbits, but they still have little probability of reaching the target. These ions receive only a fraction (approximately one-half or less) of the accelerating

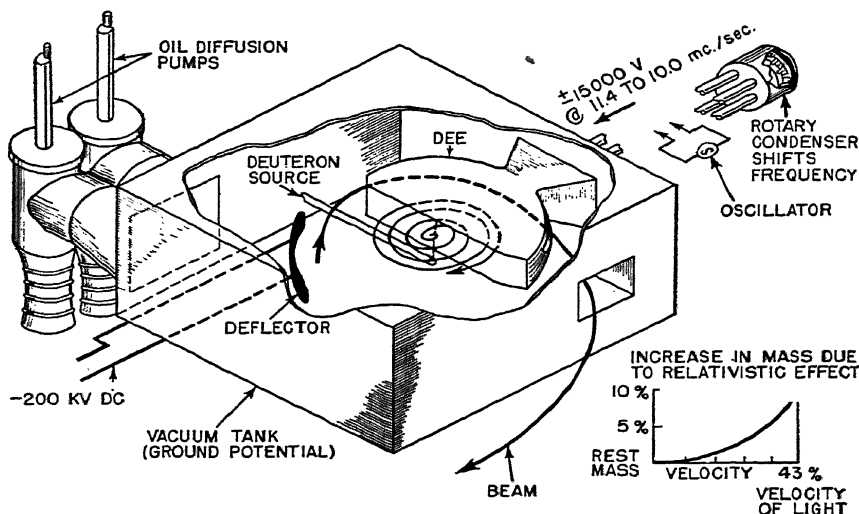


FIG. 128. Diagram of the principal components of the 184-in. cyclotron of the Radiation Laboratory, University of California. [Reprinted by permission of the Radiation Laboratory.]

voltage at each transit past a gap and must perform correspondingly more cycles; hence, they traverse a greater path length to reach the maximum radius and energy. The probability of collision with the residual gas in the tank becomes greater and, in general, such ions are either deflected by collisions to the dee walls or are knocked out of resonance with the electric field. The ions that successfully reach the target, therefore, are those which start during an interval in the electric-field cycle shortly after it has passed maximum voltage, as illustrated in Fig. 129.

For those ions which have successfully entered circular orbits three features of the cyclotron are essential to their subsequent acceleration to the maximum radius and energy: (1) stability to small oscillations superimposed on the normal spiral path; (2) the tendency of ions to remain in or near the median plane between the pole pieces; and (3) the stability of resonance between the ion rotational frequency and the oscillating electric field. The necessity for

a high order of stability is apparent from the fact that in large cyclotrons the ions will have traveled a total path length of hundreds of meters before arriving at the target where the beam should have a cross-sectional area no greater than a few square centimeters. Consequently small deviations from the normal orbit and from the correct phase with respect to the dee voltage must either be corrected while the ions are in transit or must lead to stable oscillations about the normal orbit. The reasons for the high degree of stability in cyclotron orbits is to be found in the ion focusing properties of the electric and magnetic fields and the varying amounts of energy that ions

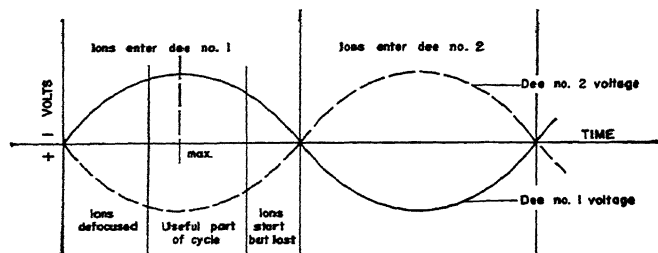


FIG. 129. Starting phase of particles in the cyclotron with respect to the radio-frequency dee voltage.

with different phase angles derive from the electric field in traversing the gaps.

Electrostatic focusing alone is important during the first part of the acceleration, before the ions have reached large radii and high energies. When an ion, that is not in or parallel to the median plane traverses the first half of the gap between dees, the electric field, because of its lenslike contour, focuses the ion toward the median plane, as shown in Fig. 130. In the second half of the gap, the symmetry in the field causes an opposite or defocusing effect. The ion, however, has increased its momentum, and the angle of deflection is then less. The deflection toward the median plane is always greater, and the net effect, therefore, is focusing. If the ion traverses the gap shortly after the peak dee voltage has been reached, the focusing effect is further augmented since the field weakens during transit and the particle is focused both by its momentum increase and also by the asymmetrical field it now experiences. The reverse process follows when transit occurs during the part of the cycle when the voltage is increasing rapidly; defocusing is enhanced and may exceed all focusing effects. The ion is then lost by collision with the dee or other structural parts after a sufficient number of periods.

Magnetic focusing fortunately becomes predominant at large radii and high energies where electrostatic focusing becomes ineffective. This effect

arises from the bowed magnetic field near the edge of the gap, providing horizontal components in the magnetic field, as shown in Fig. 131. Ions that have wandered out of the median plane are continuously forced back by interaction of their vertical velocity components with the horizontal field

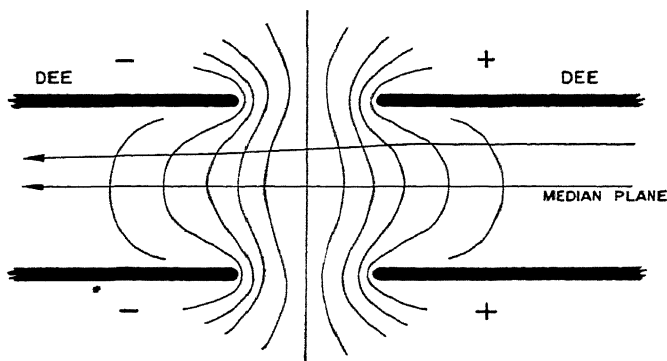


FIG. 130. Electric field focusing effect on ions in a cyclotron. The figure illustrates the potential distribution in the gap between dees. Due to the lenslike contour of the field, particles in trajectories lying outside the median plane are deflected back toward the plane.

component in the same way that the particle is constrained to move in a circular or spiral orbit by the vertical component of the field.

Both the electrostatic and magnetic focusing effects provide the requisite restoring force to ensure the stability of oscillations of the orbits about the median plane and the normal orbit. When the magnetic field possesses

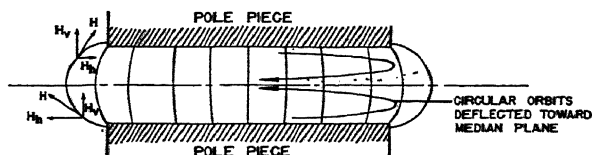


FIG. 131. Magnetic focusing effect on ion trajectories of large radius in the cyclotron. Cross-sectional view of magnetic field shows bowed lines of force near periphery which deflect particles toward median plane.

perfect radial symmetry, free oscillation can occur with both vertical and radial components and only with periods equal to or longer than the rotational period of the particle. Free oscillations in cyclotrons are neither damped nor forced once they are excited.

Oscillations in phase, *i.e.*, the angular position of the particle with respect to the gap when the dee voltage is zero, are stable for certain orbits. A particle arriving at the gap early or late is retarded or advanced accordingly

on successive transits and continues to execute simple harmonic motion about the equilibrium phase.

Azimuthal asymmetries in the magnetic field lead to forced oscillations with frequencies equal to or greater than that of the rotational frequency. Small oscillations of this form are usually stable and sometimes must be tolerated.

**21.3. Ion Source.** The ion source in a cyclotron is accurately located at the center of the magnetic field and directly above or below the gap between the dees, corresponding to the position in the median plane from which the

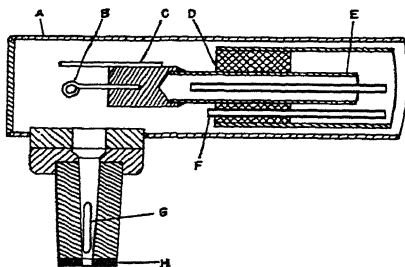


FIG. 132. Typical cyclotron ion source. A, support tube enclosing filament structure; B, helical tungsten filament; C, molybdenum or stainless-steel filament shield; D, filament lead insulator; E, filament leads and clamp with squirt tube for water circulation; G, vertical slots in cone facing dees, ionization occurs in hole through cone; H, tungsten cap on copper cone to prevent local melting due to ion bombardment from below.

greatest number of ions can be started on stable orbits. In its conventional form the ion source consists of either a helical or U-shaped tungsten filament mounted in a closed copper structure that contains a capillary outlet directed toward the median plane and parallel to the magnetic field, as shown in Fig. 132. The entire structure is supported from the tank wall by an appropriate mechanism to allow exact adjustment of its position with respect to the dees for optimum beam output. The gas to be ionized, hydrogen, deuterium, or helium, is piped into the space around the filament, and from there it leaks out through the capillary in which it is ionized by electron bombardment.

An ion cloud is then formed in the central region between the dees from which ions are pulled, each half-cycle, into alternate dees by the strong electric field. A tungsten-capped block mounted on the side of the tank opposite the source frequently serves as an anode, operating at several hundred volts positive with respect to the filament structure. Filaments have been successfully operated on direct as well as on low- and high-frequency current.

Details of the design of ion sources vary somewhat depending upon the particles to be accelerated and the particular cyclotron in which they are used. Usually the same source can be employed for protons and deuterons. Alpha-particle sources, however, are made more massive to withstand the higher rate of deterioration from ion bombardment. Water cooling of the filament leads is essential as is good thermal contact between parts of the source to prevent local melting from ion bombardment.



**21.4. Cyclotron Targets.** *a. Design Features of Targets.* A variety of target forms and techniques are employed, depending largely upon the physical properties of the bombarded material, the purpose of the bombardment, and whether the material is bombarded in the vacuum chamber of the cyclotron or in an external chamber into which the beam is deflected.

An internal target requires special consideration in its design and is limited to the comparatively few substances that are stable at elevated temperatures in a vacuum. Although a slightly higher bombardment efficiency can be obtained than is usually possible for external targets, the limitations and operational difficulties make internal targets inconvenient for routine bombardments. Nevertheless, internal targets are frequently used in one of two possible forms. In the first, the target material is mounted at the end of a probe and inserted directly into the circulating beam at its maximum radius [3]. In the second form, the target is mounted in a chamber located beyond the deflector. Probe targets are usually less satisfactory for many purposes, but they possess the advantage that considerably greater beam currents can be obtained, and the deflected beam to the target chamber can sometimes be used simultaneously although with some loss of intensity.

Bombardment in a target chamber into which the beam is deflected is the most widely practiced use of internal targets [4]. Such targets can be changed rapidly by means of a vacuum lock and, in general, engineering and operational factors are more favorable.

External targets are also bombarded in a target chamber located beyond the beam deflector but are separated from the vacuum tank by a thin aluminum window through which the beam emerges. Comparatively little difficulty is experienced with these targets since occluded gases, high vapor pressure, and decomposition in no way affect operation of the cyclotron. Such targets can be operated when necessary in an atmosphere of helium at a reduced pressure to prevent oxidation and to avoid excessive beam absorption. In addition to simplicity of operation and design of external targets, gases and liquids can be bombarded as well as most solid substances.

Practically all substances must be mounted on a water-cooled copper target plate to be bombarded, both for internal and external targets. The details of the design of a target and the choice of target material are, in general, governed by heat transfer, bonding properties, stability of the substance, bombarding efficiency, and the subsequent chemistry. The relative importance of the first three factors in the design difference of internal and external targets is chiefly one of degree, whereas the last two factors apply equally well to both types of targets.

*b. Heat Transfer.* Provision for rapid transfer of heat must be made between the target substance and the cooling system. Although as little as 25 watts may be delivered by the beam, the equivalent heat is liberated

usually in the very small volume of the target material corresponding to the beam cross section and depth of penetration. The bonding to the water-cooled copper target plate should ensure good thermal contact, and the thickness of the material should be limited to prevent high surface temperatures and local overheating due to steep thermal gradients. This is particularly important for internal targets when the material has an appreciable vapor pressure or a tendency to decompose at elevated temperatures.

*c. Target Materials and Bonding.* The most satisfactory processes for bonding materials to the copper target plate are electroplating, soldering, and brazing. They are applicable, however, only to those few metallic elements and alloys, including beryllium which can be silver soldered, that are stable under bombardment and at elevated temperature, particularly when used with internal targets. Materials that cannot be bonded by these processes are usually prepared in the form of suitable compounds or mixtures. Many of these can be fused and will adhere to a copper target plate on which V-shaped grooves have been milled in a direction parallel to the plane of the beam and to a depth corresponding roughly to the range of the particles in the target material. A thin layer of the material is spread over the grooves and the plate heated in a furnace or from the bottom with a torch until the material fuses into the grooves. Better heat transfer to the plate is obtained if the grooves are not quite filled.

The choice of compound or mixture used for this type of target is determined mainly by its binding strength and stability. For internal targets this is particularly important; it should not decompose, melt, or have a vapor pressure high enough at elevated temperatures to interfere with operation of the cyclotron or to cause loss of material. These factors are less stringent in external targets, but it is still desirable to use materials with high resistance to thermal shock and very low vapor pressure in order to reduce loss of active material.

The suitability of a substance for internal targets is difficult to predict beforehand, but several generalizations of the properties of compounds best suited for target materials have been proposed by Reid [4] and Weil [5]:

1. Ionic compounds of two elements probably will not adhere well.
2. Ionic compounds in which an ion is diatomic adhere better. If the anion is the metaform, bonding may be good.
3. Compounds that polymerize may be satisfactory.
4. Compounds that form a glassy, noncrystalline solid generally form a good bond.

A list of compounds used for internal targets is given in Table 44.

Many materials that are stable under target conditions but cannot be fused easily to the target can be bombarded in the form of powder. Targets in this form are prepared by filling a shallow recess in a water-cooled copper

block with the powder and sealing it in with a cover of thin tantalum foil [6]. It is then bombarded externally, usually in an atmosphere of helium at reduced pressure.

Both gaseous and liquid substances can be readily bombarded externally in appropriately designed containers. One end of such a container is sealed with an aluminum or tantalum foil through which the beam enters. As in solid targets, proper consideration should be given to the dissipation of heat produced in the target volume.

TABLE 44. FUSABLE COMPOUNDS USED FOR CYCLOTRON TARGETS [4,6]

Element bombarded	Target compound	Element bombarded	Target compound
B	$B_2O_3$	K	$K_2B_2O_4$
O	$Li_2B_2O_4$		KCl
	$B_2O_3$	V	$KVO_4$
F	$Li_2AlF_6$	Ge	$GeCu_2$
Na	$Na_2B_2O_4$	As	$KAsO_2$
P	$Fe_2P$	Te	$TlTeO_3$
	$Ca(PO_3)_2$		

*d. Target Efficiency.* From the point of view of beam utilization, target efficiency is judged by the proportion of the beam producing the desired reaction. An efficiency of 100 per cent is assured, therefore, only in a target consisting wholly of the isotope producing the reaction; the presence of isotopes of the same species reduces the efficiency by the amount of their total abundance compared to the isotope producing the reaction. When the target material is a compound or mixture, the efficiency is still further reduced and in a more complicated manner since the stopping power of the constituent atoms is different. Obviously, compounds containing only a small percentage of the reaction element should be avoided when efficient bombardment is sought. The volume of target material is also a factor in the efficiency when its depth is less than the useful range of the particle and its surface does not completely cover the cross section of the beam.

So far as bombardment efficiency is concerned, it would appear desirable to make the target more than necessarily massive to ensure complete beam absorption in the target material; however, other considerations make it desirable to reduce the volume of material to the minimum required for complete beam utilization. This is necessary to obtain the highest specific activity and to reduce the volume of material handled in chemical isolation of the active isotope. The surface area should correspond to the beam cross section which is usually known and reasonably stable. The depth should equal the maximum useful range of the particles in the particular target

material. The useful range, however, need not correspond to the total range of the particle in the substance. When a threshold energy for the reaction exists, that portion of the residual range in which the particle energy is less than the threshold does not contribute to the reaction and might just as well lie within the copper target plate. In estimating the target thickness, therefore, a length equal to the threshold range should be subtracted from the total range in the material. Any scattering effects in targets, particularly of heavy elements and for low energies, would tend to increase the effective thickness of the target.

The ranges for protons, deuterons, and alpha particles are given for a variety of substances in Chap. 4, but for those elements for which ranges are not given, a rough approximation, good enough for this purpose, can be obtained by extrapolating from the range-energy data for an element of nearly the same atomic number given in the graphs. If  $R_o$  is the known range of a particle at a particular energy in an element of atomic number  $Z_o$ , the range  $R$  in another element with atomic number  $Z$  is

$$R \simeq R_o \frac{Z_o N_o}{ZN}$$

where  $N_o$  = number of atoms per cc of atomic number  $Z_o$ .

$N$  = number of atoms per cc of atomic number  $Z$

When the target material is a compound or mixture, it is possible in many cases to use an average atomic weight for computing the range by extrapolation; then

$$R = \frac{R_o Z_o N_o}{N \sum_{i=1}^n f_i Z_i}$$

where  $f_i$  = fraction of atoms with atomic number  $Z_i$  in substance containing  $n$  atomic species

**21.5. Synchro-Cyclotron.** Early in the development of the cyclotron it was pointed out that the relativistic increase in mass of an accelerated particle would place a limit on the maximum energy derivable from the conventional form of cyclotron [9]. With only a few per cent increase in mass this effect becomes troublesome; the rotational frequency of the particle decreases, and it arrives at the gap later each successive cycle. The phase of the particle with respect to the fixed-frequency electric field decreases until ultimately the particle crosses the gap at zero voltage and is no longer accelerated. Although high energies can be imparted to charged particles despite their reduction in angular velocity and the accompanying phase shift, the dee voltage required to do this becomes prohibitively great.

This limitation, inherent in a combination of fixed magnetic and constant-frequency electric fields, can be removed either by slowly increasing the magnetic field or by slowly reducing the electric-field frequency to match the particle's angular velocity during the period of acceleration [10-13]. The first method is impracticable in the magnetic structure required for the cyclotron. The second alternative, however, has proved successful and has been used for the acceleration of deuterons to  $\sim 200$  mev [14,15] corresponding to a mass increase of 10 per cent. The dee-voltage frequency is slowly decreased over the period of time required by an ion starting from the center to reach the maximum radius. The per cent frequency modulation required depends on the mass of the ion and the maximum energy to which it is accelerated. Exact correspondence between the changing rotational frequency and the modulation frequency, or more correctly, their instantaneous rates of change, is not necessary since it has been shown that a slight mismatch in the two frequencies leads to oscillations about the equilibrium phase which are stable for an adiabatically changing field frequency. Precise control of the field, therefore, is not an important factor to contend with, and the field frequency need only approximate the instantaneous rotational frequency of the particle given by

$$f = \frac{f_o H}{(1 + E) H_o}$$

where  $f_o$  = resonant frequency at low energies.

$E$  = energy of particle in units of  $Mc^2$

$H_o$  = magnetic field strength at center

$H$  = magnetic field strength at radius where energy is  $E$

An example of the frequency-modulated cyclotron is shown in Figs. 127 and 128, illustrating the 184-in. cyclotron of the University of California. Modulation is accomplished with a rotating condenser mounted at the end of the dee stem [15,16].

Unlike the conventional cyclotron, the beam in this modified form must be pulsed. Ions are injected during an interval of a few microseconds only at the beginning of the modulation cycle when the electric-field frequency is in resonance with the particle's rotational frequency at low energies. Aside from frequency modulation and a pulsed beam, the characteristics and structure of the synchro-cyclotron are similar to the conventional cyclotron. The ultimate energy that can be reached is now dependent wholly on practical considerations.

Modulation of the field frequency can be accomplished by varying either the inductance or capacitance of the resonant dee circuit. Several mechanical systems have been successfully developed which provide frequency

modulation up to 30 per cent [14-16]. In all cases variable capacitors were used.

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## CHAPTER 22

### THE BETATRON

**22.1. Description.** The betatron was first successfully developed by Kerst [1,2] for the acceleration of electrons by electromagnetic induction. In principle, it is similar in operation to the common shell-type transformer, but instead of employing a secondary coil, electrons are injected into an evacuated "doughnut" to move freely in a circular path to which they are constrained by a magnetic field. A large part of the magnetic flux produced in the magnet core is made to loop through the circular orbits by an arrangement of narrow air gaps and iron cores. Thus, as the flux is increased or decreased, a tangentially induced electric field is experienced by an electron in an orbit of radius  $r$  identical to the field induced in a secondary winding.

$$E = \frac{-1}{2\pi r c} \frac{d\Phi}{dt}$$

where  $c$  = velocity of light

$\Phi$  = magnetic flux through the electronic orbit

The magnetic field  $H$  at the orbit must also increase at the same rate as the flux. If both the electron momentum and the magnetic field increase in proportion, the radius of the orbit will remain constant during the interval the electron is accelerated. The condition necessary for the maintenance of an equilibrium orbit of fixed radius  $r_o$  is given by the betatron flux condition

$$\Phi - \Phi_o = 2\pi r_o^2 H$$

where  $\Phi_o$  = initial flux when  $H = 0$

$\Phi$  = flux at any instant  $t$

$H$  = magnetic field at equilibrium orbit at same instant

The flux through the orbit must be proportional to twice the field strength at the orbit; this condition is easily realized in practice since both the linkage flux and the magnetic field at the orbit are produced in the same gap by a single magnet.

Acceleration of electrons from the initial injection energy to the maximum energy occurs in one quarter of the magnetic field cycle, either when the field is increasing or decreasing; in either case the direction of rotation is reversed. The low-frequency alternating magnetic field is produced by coils wound on the upper and lower pole pieces and, in the most efficient form, coupled to a bank of condensers of appropriate capacity to form either a

resonant or a controlled-tank circuit. A generator is coupled to the circuit through a secondary winding to compensate for losses in the iron and losses due to ohmic resistance.

A structure of laminated transformer iron is required for the yoke and pole pieces in order to reduce excessive heating and losses by induced eddy currents. Similarly, large metallic parts cannot be used in or near the region of the gap since eddy currents induced in conductors in this region cause sufficient alteration in the magnetic field to disturb the circulating electrons. The vacuum chamber or doughnut, therefore, is constructed wholly of non-conductive materials such as glass, quartz, or porcelain.

**22.2. Electron Injection and Beam Extraction.** Electrons are injected into the accelerating region from a source that is pulsed for a few microseconds

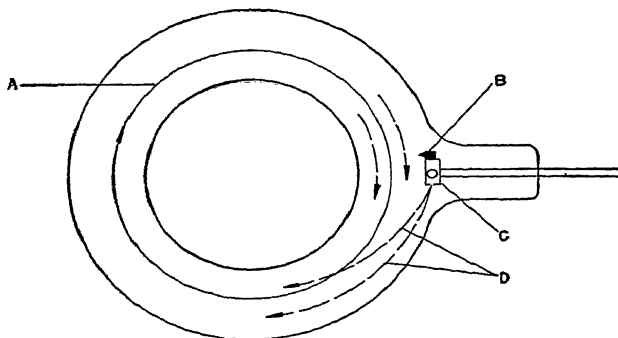


FIG. 133. Electron paths in the betatron. *A*, equilibrium orbit; *B*, tungsten target; *C*, injector containing filament and electrostatic accelerating slit; *D*, electrons leaving injector on oscillating orbits.

at the proper instant in the magnetic-field cycle. Normally the electrons are given an energy of 20 to 70 kev by electrostatic acceleration in the injector in order to provide electrons with a velocity near that of light. A comparatively simple source is used consisting of a hot cathode for emitting electrons and a slit anode through which a narrow beam of electrons is accelerated, as shown in Fig. 133.

Several possible mechanisms have been proposed for extraction of the beam when the maximum energy is attained. In all cases, the beam is deflected from the equilibrium radius either to smaller radii where it ultimately impinges on a tungsten target to be converted into x-radiation, or to larger radii to strike a target mounted on the rear of the injector. The simplest scheme for extraction requires saturation of the pole pieces just before the maximum flux through the orbit is reached. The flux conditions are then no longer maintained; the electrons gain momentum faster than the



magnetic field at the orbit increases and, hence, spiral outward until they strike the tungsten target mounted on the back of the injector.

A second and more practicable method of beam extraction alters the flux condition by the use of auxiliary coils at the pole pieces [3]. At the end of the first quarter cycle of the magnetic field these coils are energized by the discharge from a condenser bank and, depending on the polarity, the field at the equilibrium orbit is increased or decreased while the linkage flux is little altered. The electrons correspondingly spiral inward or outward to reach the target.

**22.3. Orbital Oscillations.** The existence of an equilibrium orbit when the flux condition is maintained suggests that oscillations about this orbit may also be stable. This is a property of special importance because not all electrons can be started or maintained in precisely the equilibrium orbit. If the magnetic field is radially symmetrical, the principal oscillations are vertical and radial. As shown by Kerst and Serber [4], such oscillations will have frequencies given by

$$\begin{aligned}\Omega_{\text{vert}} &= n^{1/2}\omega \\ \Omega_{\text{rad}} &= (1-n)^{1/2}\omega \\ n &= \frac{d(\ln H)}{d(\ln r)}\end{aligned}$$

where  $\omega$  = angular velocity of electrons in equilibrium orbit

Excitation of these modes of vibration accompanies injection as a result of the angular spread in the injected electrons and the radial displacement of the injector from the equilibrium orbit. Electrons leaving the injector must spiral inward to approach the stable orbit whose radius satisfies the flux condition; but, depending upon the initial angle of injection, some electrons may overshoot and others undershoot the equilibrium orbit and subsequently oscillate with simple harmonic motion about the correct radius and the median plane with a frequency usually less than that of rotation. Further excitation of these oscillations may occur during acceleration both from the effect of space charge spreading and from scattering by the residual gas in the vacuum chamber.

Two damping factors appear once oscillations are excited. As the magnetic field increases, the amplitude of oscillation is diminished in proportion to  $H^{-1/2}$ . At high energies further damping is introduced by radiation loss. For a radially symmetrical field, no forcing factors are present to maintain the amplitude of oscillations following the initial exciting pulse resulting from overshooting or scattering.

**22.4. Focusing.** Although the equilibrium radius is fixed by the flux condition, it is necessary to provide vertical magnetic focusing to keep the beam in or near the median plane during the long total path length which, in

larger instruments, is greater than 10 km. This is readily accomplished with wedge-shaped pole faces giving a magnetic field that is bowed outwardly over the region of the electron orbits, as shown in Fig. 134.

The amount of focusing that can be provided is limited by the accompanying radial decrease in magnetic field since the concurrent stability to radial

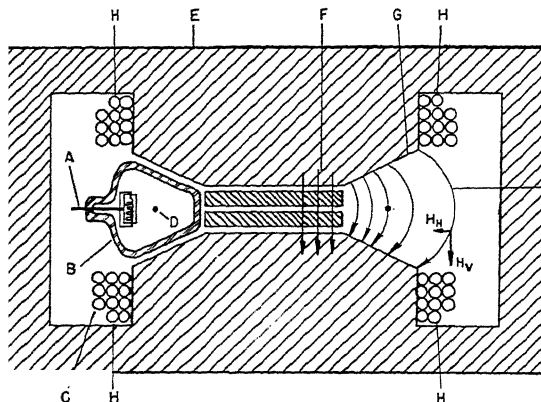


FIG. 134. Schematic diagram of betatron construction. A, electron injector; B, "doughnut" vacuum chamber; C, auxiliary coil for extracting beam; D, equilibrium orbit position; E, magnet yoke; F, flux path; G, pole faces, wedge-shaped for focusing; H, magnet energizing coils; I, bowed magnetic field in region of orbit showing horizontal component necessary for focusing.

oscillations is possible only when the restraining force of the magnetic field falls off with radius less rapidly than the centrifugal force on the electron. The magnetic field, therefore, must decrease with radius at a rate less than  $1/r$ , or in general, stable, focused orbits are obtainable when

$$H \sim \frac{1}{r^n} \quad 0 < n < 1$$

The defocusing effects of space charge and scattering are strongest at low electron velocities when, fortunately, magnetic focusing is most effective.

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## CHAPTER 23

### THE SYNCHROTRON

**23.1. Description.** The synchrotron, proposed independently by McMillan [1] and Veksler [2], employs certain properties of both the cyclotron and betatron to accelerate electrons to very high energies. Electrons, injected with a velocity near that of light with an electron gun, are first accelerated by betatron operation for which flux bars (Fig. 135) located within the orbit provide the necessary time rate of change of flux through the orbit due to the slowly increasing magnetic field. At a certain energy the flux bars become saturated, the flux remains essentially constant thereafter, and betatron operation ceases. At this time a constant radio-frequency voltage is applied to the dee and the electrons are then accelerated to high energies by cyclotron-like operation. Since electrons are injected with nearly the velocity of light, the radius increases by only a few per cent during subsequent acceleration. The magnetic field, pole pieces, and vacuum chamber therefore are confined to a narrow annular region about the stable orbit, as illustrated in Fig. 136.

A principal characteristic of the synchrotron is its "phase stability." The angular velocity of an electron traversing a circular trajectory normal to a magnetic field is

$$\omega = \frac{eH}{mc} = \frac{ecH}{E}$$

where  $e$  = electronic charge

$m$  = relativistic electronic mass

$H$  = magnetic field strength

$c$  = velocity of light

$E$  = total energy of electron, kinetic plus rest energy

As the energy and relativistic mass of the electron increases, the magnetic field at the orbit must increase correspondingly if the electron is to remain in or near resonance with the oscillating electric field applied to the dee. With a magnetic field that increases slowly during the period of acceleration, oscillations in phase are stable and tend to be damped. In effect, the phase stability causes the particles to remain in or near resonance with the electric field, making only small departures from exact resonance. This can be shown by the fact that an electron traversing the dee gap too early, *i.e.* at a time when the dee voltage is low, receives a smaller impulse than an electron

arriving late when the voltage has increased. The angular velocity of the first electron is therefore reduced and on each successive transit it will arrive at the gap later. The second electron, however, increases in angular velocity and arrives at the dee gap earlier on successive cycles. The electrons initially injected therefore tend to bunch about on equilibrium phase with

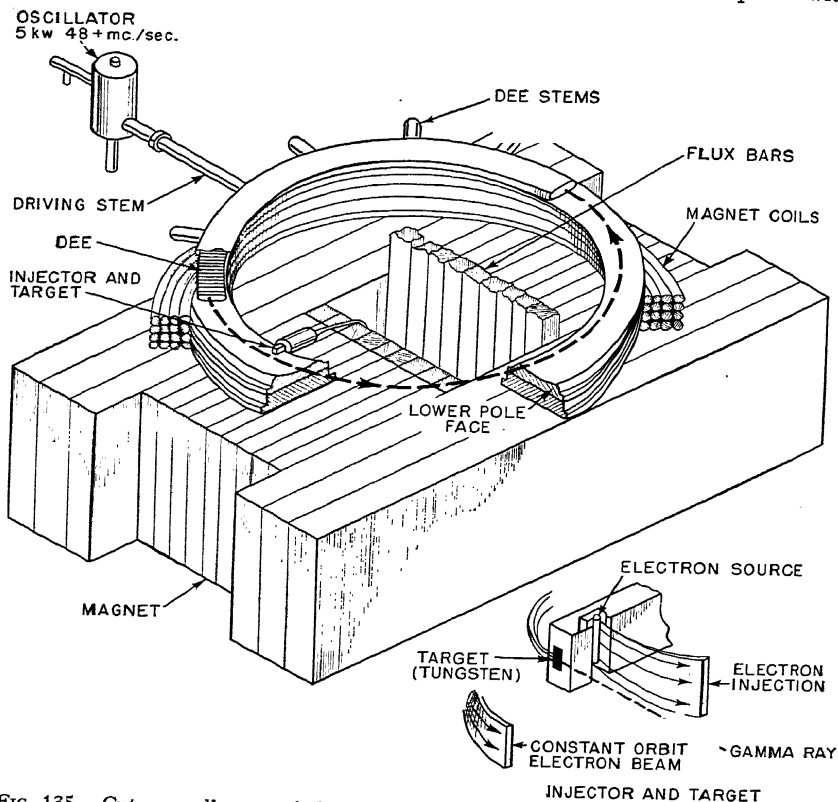


FIG. 135. Cut-away diagram of the 335-mev synchrotron in operation at the Radiation Laboratory, University of California. View shown with top half of magnet removed. [Courtesy of Atomic Energy Commission, University of California.]

respect to the electric field during the early stages of synchrotron operation. Oscillations about the equilibrium phase continue throughout acceleration, but their amplitude is diminished by both radiative loss and by the increasing magnetic field.

If the magnetic field were constant, the electrons would not gain energy

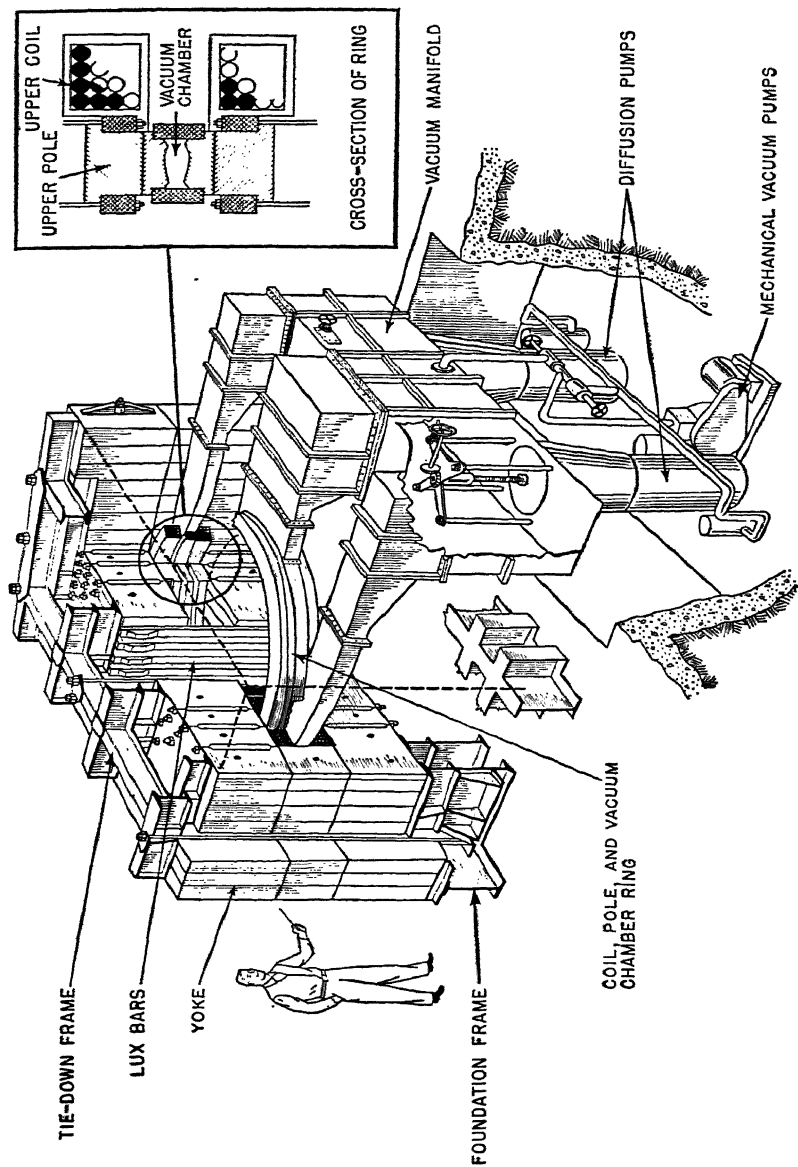


FIG. 136. Cut-away diagram showing the main structural features of the 335-mev synchrotron in operation at the Radiation Laboratory, University of California. [Courtesy of Atomic Energy Commission, University of California.]

but would maintain a constant average energy while performing phase oscillations about the equilibrium phase. However, if the magnetic field is increased adiabatically, *i.e.*, an infinitesimal increase in  $H$  during one cycle of the particles, the phase oscillations will still remain stable but the circulating electron group as a whole must receive a net increase in energy from the electric field at each transit of the gap if the angular frequency is to remain constant. The ultimate energy is not limited by the radius as in the cyclotron, since here it remains nearly constant, but rather by radiative losses from the electrons at very high energies. An energy can, in principle, be reached for which the energy loss by radiation just equals the energy derived per cycle from the electric field.

In principle, the structure of the synchrotron magnet is similar to the core of a large transformer. The yoke is built up from thin laminated plates of iron to reduce excessive heating and loss from eddy currents. The annular pole pieces similarly are constructed of thin laminations bonded with a non-conducting material and shaped to give the requisite bowed field required for focusing. The varying magnetic field is produced by coils wound on the upper and lower pole pieces and energized at the desired frequency by discharges from banks of condensers.

Structural details of a synchrotron built for the Atomic Energy Commission at the University of California are shown in Figs. 135 and 136.

**23.2. Motion of Particles.** *a. Synchronous Orbits.* The condition for a synchronous orbit is satisfied when the electron rotates in resonance with the electric field at an angular velocity given [1,2] by

$$\omega_s = \frac{eH}{m_s c} = \frac{ecH}{E_s}$$

Alternatively, this gives the equilibrium energy at a particular value of  $H$ . The angular position of an electron with respect to the gap when the dee voltage is zero is referred as the phase  $\varphi$ . In the synchronous orbit, the phase is constant and results in an energy gain per turn of  $eV \sin \varphi$ , where  $V$  is the peak radio-frequency dee voltage. The equilibrium or synchronous phase, which can be determined from the conservation of energy, is given [1,3] by the expression

$$eV \sin \varphi_s = \Delta E_s + L_s - 2\pi r_s V_1$$

where  $\Delta E_s = \frac{2\pi}{\omega^2} \frac{ec}{dt} =$  energy gain per turn

$L =$  radiation loss per turn

$r = (E^2 - m^2 c^2)/300H =$  equilibrium radius

$V_1 =$  voltage due to changing magnetic flux. This contribution is usually small after synchrotron operation has started

*b. Phase Oscillations.* Electrons that were not started in synchronous orbits or that arrive at the gap at a time different than for the equilibrium phase are retarded or advanced on successive transits and thus continue to execute stable oscillations in phase as well as in radius and energy about the synchronous values. The equation of motion of the phase  $\varphi$  is given [1,3,4] by the relations

$$\frac{d}{dt} \left( \frac{E_s}{\omega_s^2 K} \right) + \frac{r_s L'_s c^2 \varphi}{2\pi(1-n)K v_s^2 \omega_s} + \frac{eV}{2\pi} \sin \varphi = \frac{eV}{2\pi} \sin \varphi_s$$

where

$$K = 1 + \frac{n}{(1-n)} \frac{c^2}{v_s^2} \sim \frac{1}{1-n}$$

$$n = - \frac{d(\ln H)}{d(\ln r)}$$

$$L'_s = \frac{dL_s}{dr}$$

$$v_s \sim c = \text{velocity of light}$$

For oscillations of small amplitude, the energy also oscillates about the equilibrium value by the amount

$$\Delta E = - \left( \frac{eV E_s \cos \varphi}{2\pi K} \right)^{1/2} (\varphi_m - \varphi_s)$$

where  $\varphi_m$  = maximum phase amplitude

The corresponding change in radius is given by

$$\Delta r = \frac{c^2}{v_s^2} \frac{r_s}{(1-n)} \frac{\Delta E}{E_s}$$

In general, the amplitudes  $\varphi_m - \varphi_s$ ,  $\Delta E$  and  $\Delta r$  vary as

$$\varphi_m - \varphi_s \sim [(1-n)VE_s]^{-1/4}$$

$$\Delta E \sim E_s \left[ (1-n) \frac{V}{E_s} \right]^{1/4}$$

$$\Delta r \sim r_s \left[ \frac{V}{(1-n)^2 E_s^3} \right]^{-1/4}$$

The frequency of phase oscillation is small compared to the orbital frequency of the electrons. From the equation for the phase, its frequency is found [3] to be

$$\Omega = \omega_s \left( \frac{eVK \cos \varphi_s}{2\pi E_s} \right)^{1/2} \left[ 1 - \frac{1}{16} (\varphi_m - \varphi)^2 \right]$$

These oscillations were shown to be stable for adiabatic changes in the magnetic field [3].

*c. Orbital Oscillations.* Both vertical and radial oscillations in the orbits may occur with frequencies comparable to the rotational frequency of the electron. Calculations of these frequencies [5] gives values of

$$\begin{aligned}\Omega_{\text{vert}} &= n^{1/2}\omega \\ \Omega_{\text{rad}} &= (1 - n)^{1/2}\omega \\ n &= \frac{d(\ln H)}{d(\ln r)}\end{aligned}$$

which are damped in part by the increasing magnetic field and in part by radiation loss at high energies.

**23.3. Energy Loss by Radiation.** Electrons moving in circular orbits with velocities very close to the velocity of light lose energy by radiation arising from radial acceleration. The energy loss per turn due to incoherent radiation is given by

$$L = \frac{4\pi e^2}{3r} \left( \frac{E}{mc^2} \right)^4$$

Additional radiation resulting from orbital and phase accelerations has the effect of damping such oscillations. For phase oscillation the decrease in phase amplitude per turn due to radiation damping is

$$\Delta(\varphi_m - \varphi_s) = \frac{1}{2} (3 - 4n) \frac{L_s}{E_s}$$

**23.4. Synchrotron Operation.** Shortly after the magnetic field cycle begins, electrons are injected into the accelerating region with an energy of approximately 60 kev. By betatron action (radio-frequency field off) they are then accelerated by the constant torque of the induced field to an energy of the order of 2 mev and a velocity of approximately 0.99 c. By this means the orbits are rapidly brought clear of the injector and to a radius that changes very little on subsequent acceleration. The transition from betatron to synchrotron operation takes place when  $E_\beta = E_s$  or  $r_\beta = r_s$ . During the period of transition the electrons increase in angular velocity until their period matches that of the radio-frequency field which, during this time, is slowly increasing in amplitude to its peak value. Those electrons lying within the proper phase angles are then "locked" into synchronism and gradually bunched. Betatron action effectively ceases with saturation of the flux bars, and the electrons thereafter rapidly derive energy from the radio-frequency field.

Despite the bunching effect an energy spread persists because of the finite duration of the injection time. Assuming that the injection interval corresponds to the time required for an electron to increase from radius  $r_o$  to  $r_\beta$ , the energy spread can be calculated [3] from the expression

$$\delta E = (1 - n)(2mc^2T) \frac{r_\phi - r_\beta}{r_\beta}$$

where  $T$  = kinetic energy of electrons at injection

$r_o$  = injector radius

$r_\beta$  = betatron equilibrium radius



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TABLE 45. PHYSICAL CONSTANTS

The following is a partial list of the physical constants reported by R. T. Birge, *Rev. Mod. Phys.*, **13**, 283 (1941).

Atomic weight of electron:

1. Physical scale	$A_e$	$= (5.4862 \pm 0.0017) \times 10^{-4}$
2. Chemical scale	$A_e$	$= (5.4847 \pm 0.0017) \times 10^{-4}$
Avogadro number	$N_o$	$= (6.0228 \pm 0.0011) \times 10^{23}$ per mole
Bohr magneton	$\mu$	$= (0.9273 \pm 0.0003) \times 10^{-20}$ erg/gauss
Bohr radius	$R$	$= 0.53 \times 10^{-8}$ cm
Boltzman constant	$k$	$= (1.38047 \pm 0.00026) \times 10^{-16}$ erg/deg
Classical electronic radius	$R_e$	$= 2.807 \times 10^{-13}$ cm
Compton shift at $90^\circ$	$h/m_o c$	$= (0.024265 \pm 0.000005) \times 10^{-8}$ cm
Density of Hg ( $0^\circ\text{C}$ , standard atmosphere)	$d_o$	$= 13.59504 \pm 0.00005$ gm/cm <sup>3</sup>
Electronic charge	$e$	$= (1.60203 \pm 0.00034) \times 10^{-20}$ abs emu $= (4.8025 \pm 0.0010) \times 10^{-10}$ abs esu
Electronic mass	$m$	$= (9.1066 \pm 0.0032) \times 10^{-28}$ gm
Faraday constant:		
1. Chemical scale	$F$	$= 96,501 \pm 10$ int coulomb/gm-equiv $= 96,487 \pm 10$ abs coulomb/gm-equiv $= 9,648.7 \pm 1.$ abs emu/gm-equiv
2. Physical scale	$F$	$= 96,514 \pm 10$ abs coulomb/gm-equiv $= 9,651.4 \pm 1.0$ abs emu/gm-equiv
Fine structure constant	$\alpha$	$= (7.2976 \pm 0.0008) \times 10^{-3}$
Gas constant	$R_o$	$= (8.31436 \pm 0.00038) \times 10^7$ erg/deg/mole
Gravitational constant	$g$	$= (6.670 \pm 0.005) \times 10^{-8}$ dyne cm <sup>2</sup> /gm <sup>2</sup>
Hydrogen atom mass	$M_H$	$= (1.67339 \pm 0.00031) \times 10^{-24}$ gm
Loschmidt number ( $0^\circ\text{C}$ , standard atmosphere)	$L$	$= (2.6870 \pm 0.0005) \times 10^{19}$ per atmos/cm <sup>3</sup>
Mass of unit atomic weight ( $1/N_o$ )	$M_o$	$= (1.66035 \pm 0.00031) \times 10^{-24}$ gm
Maximum density of water	$d_{\max}$	$= 0.999972 \pm 0.000002$ gm/cm <sup>3</sup>
Planck constant	$h$	$= (6.624 \pm 0.002) \times 10^{-27}$ erg/sec
Proton mass	$M_p$	$= (1.67248 \pm 0.00031) \times 10^{-24}$ gm
Ratio physical to chemical atomic weight scales	$r$	$= 1.000272 \pm 0.000005$
Ratio proton mass to electronic	$M_p/m$	$= 1836.5 \pm 0.5$
Rest energy of electron	$mc^2$	$= 0.5107$ mev
Specific electronic charge	$e/m$	$= (1.7592 \pm 0.0005) \times 10^7$ abs emu/gm $= (5.2736 \pm 0.0015) \times 10^{17}$ abs esu/gm
Standard atmosphere	$A_o$	$= (1.013246 \pm 0.000004) \times 10^6$ dyne/cm <sup>2</sup> /atm.
Velocity of light	$c$	$= (2.99776 \pm 0.00004) \times 10^{10}$ cm/sec



PART III

Biological and Medical Applications  
of Isotopes



## CHAPTER 24

### GENERAL CRITIQUE OF THE BIOLOGICAL APPLICATION OF ISOTOPES

ELLSWORTH C. DOUGHERTY

**24.1. Introduction.** The object of Part III of the present volume is to supply information on the significance of stable isotopes and artificial radioactive isotopes in biology and medicine. This, of course, nowhere nearly exhausts the application of physical phenomena to biological and medical research. However, it is not proposed that this section be a comprehensive review of biophysics. It is rather designed as a reference source for biologists who propose to employ isotopes as tools in their work. To that end the biological literature in which the use of isotopes is recorded has been comprehensively surveyed and a bibliography prepared (initially in collaboration with M. C. Fishler), which is appended as Chap. 30. From the literature representative studies that illustrate the major applications of the isotopes already used have been selected for purposes of brief discussion. Tables of these isotopes (Tables 46 and 47) have been prepared, which list their major applications to date. A further table (Table 48) lists those isotopes which, on the basis of present evidence, have potential usefulness in biological and medical work. Part III is subdivided as follows: (1) the introductory chapter in which is presented a critique of the biological and medical application of isotopes; (2) several chapters (Chaps. 25 to 28) in which the elements, arrayed generally according to their biological significance, are considered and the use of their isotopes is discussed; (3) a chapter (29) on isotopes in medical therapy and diagnosis; and (4) the bibliography (Chap. 30).

It is intended that, guided by the general remarks of Chap. 24, the investigator who desires to use an isotope to solve a given problem will be able, with the use of Tables 46, 47, and 48, the discussions of Chaps. 25 to 29, the bibliography, and relevant chapters in Parts I and II of the present volume, to determine whether an appropriate isotope may exist for this problem. If in practice this aim is realized, the author will consider the purpose of his section fulfilled.

Consideration of the isotopes of the three naturally radioactive series has been largely omitted here for two principal reasons: (1) because the literature is extensive and voluminous, extending back to the beginning of this century and thus imposing an inordinate expenditure of time and effort

for an adequate survey; and (2) and more importantly, because a great volume of work, only partly declassified as yet, was carried out during the war by the Manhattan District Project (now U. S. Atomic Energy Commission) and will appear in the extensive series of publications planned by the Atomic Energy Commission. It therefore seems premature at this time to discuss and criticize the literature on the natural radioactive series.

The bibliography purports to include all publications through 1947 dealing with stable and artificial radioactive isotopes as tracers in biology and medicine and with the effects and differential behavior of isotopes in biological systems; papers describing chemical syntheses or physical measurements have not been included unless they also give data on biological application. There is an introductory list of general references (Gen); otherwise, the references are grouped according to element, listed alphabetically. Under each element references are in turn listed alphabetically according to author, and each is assigned the symbol of its element and a serial number. In Chaps. 24 to 29 citations are made by use of these designations (*e.g.*, Nall, 56). Papers subsequent to the end of 1947 have been specifically omitted except for those of particular importance (*e.g.*, reviews) and those in which the use of an isotope for the first time is recorded; it may be pointed out, however, that *Biological Abstracts* now includes (starting with Vol. 21, No. 7, August-September, 1947) a separate section entitled "Radioactive and Heavy Isotopes," which contains abstracts of papers on the use of isotopes and cross-references to pertinent abstracts in other sections. Furthermore, an abstracting section covering a good part of current radiobiological and isotope literature (mainly the American) is to be found in the new journal *Nucleonics*. Finally, the U. S. Atomic Energy Commission now issues a series entitled *Nuclear Science Abstracts* which includes sections dealing with biophysics, biology, and medicine.

The particular importance of isotopes in biology is related to the fact that, for most of those elements which are constituents of protoplasm, artificial radioactive species can be substituted for normally occurring homologues and used as "tracers" of metabolism and other physiological processes and that, in the case of many elements, enriched amounts of the rarer (or even the more abundant) stable species can be introduced into biological systems and also used as tracers. In addition the fate and behavior, in living systems, of minute amounts of elements and compounds not regarded as essential to life can be followed, as well as the biological performance of all sorts of labeled substances and entities normal and abnormal to an organism. Thus, certain absolute or relative properties of various atomic nuclei permit the biological application of isotopes as tracers.

Of considerable importance also is the study of isotopic behavior and effects *per se*. This is particularly true of radioactive species in connection with

the effects of their ionizing radiations. Again it is physical properties of atomic nuclei that are basic to the phenomena studied.

The biological application of isotopes can be conveniently classified as follows:

- I. Use as labeling agents ("tracers") of normal and abnormal physiology
  - A. As tracers of metabolism
    1. Of intrinsic constituents of the organism
      - a. Water
      - b. Minerals, including trace elements
      - c. Organic substances
    2. Of extrinsic substances (abnormal to the organism in form or amount)
      - a. In pharmacological studies
      - b. In toxicological studies
  - B. As tracers of nonmetabolic processes
    1. Involving cells and other intrinsic complex constituents
    2. Involving foreign substances
      - a. Dissolved gases
      - b. Colloids and particles of greater than colloidal size
      - c. Foreign cells and organisms
- II. Use in the study of the behavior and effects of isotopes per se
  - A. Differential fates of isotopes
  - B. Effects of isotopes
    1. Of abnormal concentrations
    2. Of radiation (including radiotherapy)
- III. Use as tools for biochemical analysis

**24.2. Survey of Useful Isotopes.** The 102 isotopes that have been used in biological and medical studies as tracers or for purposes of irradiating biological systems belong to 60 of the 96 known elements and are listed in Tables 46 and 47. Tracer studies in biological systems have been carried out for 55 (87 isotopes) of these 60 elements and are listed in Table 46. Isotopes of the remaining 5 elements (as well as a few of the other 55) have found use in irradiating organisms, tissues, or cells; but of these elements, two (thallium and actinium) are represented only because they have isotopes in the natural radioactive series, which thus contribute to the biological effects of the radiation from radium and radon samples. Nontracer studies are given in Table 47.

In Table 48 are listed a large number of potentially useful isotopes for which no published reports on biological tracer application have as yet appeared. These include promising species belonging to elements represented in Table 46 and also species of the 41 elements for which no tracer studies have been reported. This listing is not meant to imply that all known

isotopes omitted from the list will have no biological application. It is too early to predict what will and will not be practical in the tracer and radiobiological work of the future. In general, isotopes have been selected from the Seaborg chart on the basis of half-life, with the assumption that species with long half-lives (weeks to years) will be of most general value but that shorter lived species (hours, or even minutes, to days) may be desirable in certain cases, as, for example, for tracer and therapeutic uses in humans. Some of the isotopes in Table 48 may never find biological application by reason of some practical difficulty, as discussed in Sec. 24.3 *d*.

There are now known over 700 radioactive isotopes, of which, however, only about 233 have properties suggesting value in biological research. A total of 92 radioactive isotopes (of 56 elements) have been used in biological and medical studies, 82 (of 54 elements) of them in tracer studies. Thus, somewhat less than half of the potentially applicable species have found use in biological experimentation. Unquestionably, many, if not most, of those as yet unemployed will find experimental application in the near future. The majority of studies with radioactive isotopes have been in the field of tracer work. A number of studies, however, have been on the biological effects of radiation; and a few radioisotopes now have well-established applications in clinical medicine.

Nine stable isotopes have been applied to biological problems, and the use of a tenth ( $H^1$  as proton beams) has been suggested. Most work with stable isotopes has been with the rarer species of elements existing in more than one stable form. Of 278 known stable isotopes, 193 are less abundant species of the 60 elements having two or more naturally occurring isotopes; but of the 193, only 7 have known biological applications. Most of the work with these rarer species has been in tracer studies. However, not all the rarer stable species so far used have been applied in this manner, for certain isotopes have peculiar properties that have made possible, or suggest, their use in radiation studies. For example, both lithium and boron have rarer isotopes that split when bombarded with accelerated particles. The specific ionization of the radiation released is considerably greater than that attending the accelerated particles themselves. Thus, selective localization of lithium and boron compounds has made possible selective radiation of certain tissues. However, these properties are not necessarily restricted to the rarer species of the elements concerned. Beryllium, consisting of a single isotope ( $Be^9$ ), has given off fast neutrons when bombarded with protons or deuterons; the resulting neutron beams have in turn been used for experimental therapeutic purposes. Alpha particles, the nuclei of ordinary helium ( $\alpha$  or  $He^4$ ), and deuterons, the nuclei of heavy hydrogen ( $d$  of  $H^2$ ), have been used to irradiate living cells. Finally, the more common of the two isotopes of hydrogen



(H), *i.e.*, protium ( $p$  or  $H^1$ ), has been suggested for a possible therapeutic role when accelerated to the high energies of the new giant cyclotrons.

From the foregoing discussion it may be seen that radioactive and stable isotopes have applications both in tracer problems and in the study of radiation effects. No exclusive distinction can thus be made between them on either basis. However, for the most part radioactive isotopes are more versatile and, except in a few cases, are the important tracers. By reason of their great variety of radioactive properties, they also offer far more diversified techniques for study of radiation effects.

**24.3. Isotopes as Tracers.** Hevesy [Gen 76] has recently made an exhaustive and valuable survey of the literature on the tracer application of radioactive isotopes in biology. No such recent study exists for stable species. Since isotopes in biological tracer work are tools for the elucidation of physiological processes and for biochemical analyses rather than objects of study in themselves, a somewhat artificial separation of isotopic and nonisotopic studies of the same phenomena would be effected by a perpetuation of Hevesy's comprehensive approach. In reviewing tracer applications, the author has sought rather to cite for the isotopes of each element enough representative studies to illustrate their known range of usefulness; to these can be added in many cases speculation as to additional applications. This approach is essentially followed here in the chapters on individual elements and groups of elements.

Radioactive species that are adaptable to biological studies exist for most elements. Exceptions to this include He, Li, B, and Ne, of which the known unstable forms are so short-lived (40 sec or less in half-life) that it seems unlikely that they will ever find biological application. Certain other elements are greatly limited in this regard by the shortness of half-life of their longest lived radioactive isotopes; these include N, O, Mg, and Al, for the isotopes of which the half-lives are 10.2 min or less. Fluorine and silicon are somewhat unsatisfactory as their best isotopes have half-lives of only about 2 hr and 3 hr, respectively. The longest lived known isotope of Fa has a half-life of only 21 min. But, aside from these 11 elements, all of which are of low atomic weight except francium, the other 85 of the periodic system have radioisotopes of reasonably or very convenient half-lives for biological work.

The use of stable isotopes as tracers for the 60 elements of which more than one species exists is limited by two factors: (1) the expense and technical difficulties involved in concentrating a given species from the isotopic mixture characteristic of an element having more than one stable nuclear form; and (2) the ready availability, convenience, and as a rule, greater accuracy of application of at least one radioactive species for most elements. Only in

the case of five elements—all of light atomic weight—have practical methods been long developed for concentrating the less abundant isotope for tracer studies, namely, for H, C, N, O, and S. For H, C, and S, convenient radioactive species also exist; but successful tracing of N and O must depend in the vast preponderance of cases on the rarer stable forms. For similar reasons, if much tracing with Mg is ever to be developed, it would appear likely that some extension in stable isotope tracing must be made. The same considerations may well apply to He, Li, B, Ne, and probably Si. Aluminum is a particularly unfortunate element from this point of view inasmuch as it exists in only one stable form and has no known radioactive species of longer half-life than 6.7 min.

Recently the U. S. Atomic Energy Commission has announced the availability of isotopic mixtures for some 29 elements enriched with rarer species, primarily by electromagnetic separation (and more recently, of a thirtieth—He). These include isotopes of O and Mg. The extension of stable isotopic tracing to heavier elements, for which isotopic separation is increasingly difficult and for which convenient radioactive species are known, would seem to depend upon the desirability of entirely eliminating the effects of the radiation associated with the radioactive species of many of these elements or, in some cases, upon the need for multiple labels of a given element. So far there is little evidence that the former will prove a practical necessity; it may well be, however, that even the slight amount of radiation accompanying tracer doses will be shown, especially in the case of long-lived isotopes, to have a deleterious effect, particularly from the genetic aspect, on living organisms. Multiple labels for such elements as Ca and Fe may well prove of great value; if so, stable species will very likely play a role.

*a. Metabolic Tracing.* In the realm of metabolic tracing, the role, in normal and pathological states, of elements and compounds normal to the organism is unquestionably the most vital aspect. We deal here with both essential and presumably nonessential elements. In the former category are elements that range from those making up the major organic and inorganic constituents of protoplasm (H, C, N, O, Mg, F, S, and usually, K and Ca) to those required by all living substance in minute quantities (Fe and possibly also Mn and Zn—the so-called micronutrient, or trace, elements). The need of certain organisms for additional macronutrient elements (Na and Cl) is well known; these, together with various micronutrient elements and others “normal” to the protoplasm and protoplasmic products of various organisms embrace the greater part of the periodic system. Many presumably unessential elements usually occur in exceedingly minute amounts. Some of these, however, may eventually prove to be essential micronutrients.

Actually or potentially useful isotopic labels are known for all elements shown to be essential to living organisms (except possibly aluminum).

Universally accepted as essential to all life are the elements H, C, N, O, Mg, F, S, and Fe. Although K and Ca are usually regarded as essential metabolites, there is some evidence that certain bacteria and blue algae, for example, may not require one or the other. In addition, various elements have been shown to be unquestionably essential to certain plants or animals, and at least some of these probably are universal micronutrient requirements of living systems on our planet. These elements include B, Na, Al, Si, Cl, V, Mn, Co, Cu, Zn, Mo, and I. There is also rather good evidence that certain plants require Ga, Cb, and W, and certain animals, Ni and Br. Other elements that have been postulated as essential micronutrients for certain forms are Li, F, Cr, Rb, Sr, and Sn. It is quite possible, therefore, that some 30 or more of the 96 known elements are essential to various forms of living organisms.

So far it has been the metabolism of the macronutrient elements and of two micronutrients, Fe and I (in the higher vertebrates), that have been intensively studied with tracers. Only certain of the micronutrients have been subjected to the tracer approach, including, aside from Fe and I, the elements Mn, Co, Cu, Zn, and Mo. The important micronutrient B in plants is still to be investigated, also such probable micronutrients as Ca, Cb, and W in plants and Ni in animals.

The use of isotopic tracers has already divulged a tremendous store of data on the metabolism of essential elements. In many cases, as with much of the intermediary metabolism of organic compounds and the dynamics of electrolytes in body fluids, this information would otherwise have been quite unattainable. In the study of the role of micronutrients, isotopic tracer techniques will very likely supply us with an understanding of mechanisms now poorly or scarcely at all understood, and it may well be that these techniques can, in addition, give us evidence for the essential role of other elements not now definitely known to play such a role in any living process.

Studies with trace amounts have been carried out for a number of elements not known to be essential to life. Where these elements are normally present in minute amounts in an organism, it is possible to elucidate their behavior in the "normal" metabolism of that organism.

The study of the metabolic fate of elements and compounds abnormal to the body in form or amount has also been carried out for a large number of substances. Such investigations have either a toxicological or a pharmacological basis, or both.

*b. Nonmetabolic Tracing.* Under nonmetabolic tracing may be included study of the behavior and fate of normal body constituents above the purely biochemical level, *e.g.*, erythrocytes; and of foreign substances, *e.g.*, bacterial products and inert gases.

In some cases, it is impossible to draw a clear distinction between metabolic

and nonmetabolic tracing because the labeling isotope for the latter type of experiment often also has a metabolic role. For example, the labeling of erythrocytes with radioiron has been used to study the movement and mixing of blood ( $\text{Fe}^{59}$ ,  $\text{Fe}^{55}$ , etc.); however, the iron of labeled hemoglobin has an active role in the metabolic pool, and for long-term experiments the turnover of hemoglobin iron must be considered in any experiment with labeled erythrocytes. Similarly, in studying the movement of labeled bacteria and other foreign cells, the metabolic interchange of the labeling atoms between these cells and their host environment must be considered.

*c. Theory and Techniques of Tracing Physiological Processes.* The fundamental techniques of isotopic tracing in biology are directed toward the answering of two main questions: (1) What path or paths does a labeled material take, qualitatively and quantitatively, in moving and changing within a biological system? (2) At what rate do such processes occur? In addition, isotopes may be used in biochemical analysis by the isotope dilution technique (as already described in Sec. 15.2).

The first of these questions can in some cases be partly or even entirely answered by other than isotopic techniques. For example, at the metabolic level the tracing of an abnormal substance, or of abnormal amounts of a normal constituent, can be carried out in many cases by microchemical or even macrochemical procedures. Nevertheless, the use of isotopes even in these cases permits of greater accuracy and in many cases much greater simplicity of experimental procedure. For the study of normal metabolism, moreover, nonisotopic techniques offer at best a crude approximation of what isotopic procedures can clearly reveal. Though chemical analysis of parts of a biological system can demonstrate in large outline what the constituents are at any given time, they can tell relatively little as to the exact pattern of movement by which those constituents have become incorporated into the system. With tracers, such processes as the permeability of substances and the transport of microconstituents can be readily revealed.

The study of rates of biological processes is even less satisfactory if restricted to nonisotopic techniques. In fact only those rates can be conveniently studied nonisotopically which deal either with the production of end products or with changes in the absolute amounts or relative proportions of given constituents in a biological system. In one vital aspect, the functioning of biological systems can be satisfactorily studied only with isotopes; this is the pattern and the rate of change, or turnover rate, of intermediary substances in dynamic equilibrium in a biological system. Even though we might know by other means all the substances that enter a system and all the substances that leave it, we cannot determine merely by chemical means all the intermediate steps and, in particular, the rates at which the transformations involved are occurring. However, with isotopes as labeling

agents it is possible to put molecular groupings into a system such that they behave essentially as do similar groups already present, but are nevertheless distinguishable by virtue of slight mass differences in the case of stable isotopes or of radioactive decay phenomena in the case of radioactive isotopes.

The use of an isotope as a tracer for any given purpose involves (1) its availability in a suitable form or its incorporation into the material to be labeled; (2) its administration in a suitable manner to the biological system to be studied; (3) its recovery and measurement, or in some cases its measurement directly *in vivo*.

With radioisotopes, all these steps must be carried out with precautions against the overexposure of personnel to ionizing radiation. This involves avoidance not only of irradiation at the time of experimentation but also of contamination by, or assimilation of, radioactive materials, and hence continued irradiation. Where human beings are the biological systems studied, care must also be taken against exceeding the safe tracer dose—usually given as 0.1 rep of total body irradiation per day. Safe handling of radioactive isotopes has already been discussed in Chap. 19.

The preparation of isotopically labeled substances involves the conservation of valuable materials. Therefore, special synthetic processes, in which yields are as high as possible, often must be evolved. In some cases it is possible to obtain labeled materials through biosynthetic processes; in such cases an isotope is introduced into a living system and the desired labeled material isolated later.

The administration of labeled tracer substances presents no unusual features other than those associated with health protection against irradiation in case of radioisotopes.

The measurement of isotopes involves a number of specialized techniques. Quite different procedures must be applied to stable as opposed to radioactive species. At the present time stable isotopes may be measured only in the mass spectrometer except for deuterium and heavy oxygen, for which additional techniques are available. The latter depend either on measurements of density or on measurements of refractive index of highly purified mixtures of water and deuterium oxide ("heavy water") (*see* Chaps. 8 and 9 on stable-isotope measurement).

The radioactive isotopes may be detected or measured by three principal techniques: *in vitro*, *in vivo*, and autoradiographic. The *in vitro* and *in vivo* methods involve the use of the Geiger-Müller counter, an electroscopic counter, or an ionization chamber; the autoradiographic method, the use of a sensitive photographic emulsion.

The *in vitro* techniques of measurement are carried out on samples removed from the biological system under study and are highly varied in procedure. The choice of method depends upon the manner in which a given isotope

must be prepared for effective counting (see Chaps. 10 to 12 on radiation counters, and Chaps. 13, 17, and 18 on the standardization and preparation of samples for measurement).

The *in vivo* method is characterized by measurement of a radioisotope in the intact system. This may be accomplished only where the emanations of an isotope are sufficiently penetrating to permit the approximation of a counter to the experimental subject and the detection of particles of nuclear disintegration originating within the latter. Gamma-ray emitters and high-energy beta-particle emitters can be studied by this method. Most measurements carried out in this way are largely qualitative, although with a precise understanding of the geometry of the counting arrangement and of the distribution of the isotope within the system, accurate quantitative measurements are possible.

The autoradiographic technique is also largely qualitative. It involves the approximation of a biological sample to a photographic film sensitive to the radiations of the isotope used (see Chap. 14). The resulting pattern on the developed film illustrates the distribution and, to a certain degree, the differential concentration of the isotope on the surface of the sample and, to a greater or lesser extent, depending upon the thickness of the sample and the energy of the radiated particles, the distribution and concentration in the parts of the sample beneath its surface. The most satisfactory autoradiographs are those made from very thin samples, *e.g.*, sections of tissues or organs. Techniques have recently been developed for the simultaneous mounting of a tissue section and a sensitive gelatin layer on a microscope slide [I13,26]. The film may be developed after an appropriate interval and the section stained; the result is a stained section superimposed on its radioautograph (or vice versa) from which a precise picture of the isotope distribution can be determined.

**24.4. Differential Behavior and Effects of Isotopes.** So far there have been few demonstrations that the different stable isotopic species of a given element behave differently in living systems. However, in the case of hydrogen, the differential behavior of deuterium oxide (heavy water) has been demonstrated to have toxic effects on organisms from bacteria to mice [D31]. Evidence also has been accumulated for the differential use of carbon and oxygen isotopes by living systems [C90,O7].

Most of the radioactive isotopes in use are of sufficiently high molecular weight so that no differential effect due to the difference between their masses and those of their normally occurring stable analogues is to be expected. However, tritium will most certainly prove an exception to this inasmuch as the difference between its behavior and that of ordinary hydrogen will be greater than that already shown between deuterium and protium. The decay pattern of any given isotope is, of course, unaffected by its participation

in a biological system. However, its radioactivity can have decided effects when the isotope is present in sufficiently high concentration. In terms of the effect of ionizing radiation, its effect may be profound.

Radioactive isotopes have been used in a number of studies on the effects of ionizing radiation on biological systems. In particular, interest has been focused on those isotopes which can be administered in a form that will be selectively localized in specific parts of such systems. The basis for the use of radioisotopes in therapy lies in the phenomena of differential concentration. Dosimetry of radioisotopes has already been discussed in Chap. 16.

## CHAPTER 25

# ELEMENTS CONSTITUTING MAJOR ORGANIC METABOLITES

ELLSWORTH C. DOUGHERTY

**25.1. Introduction.** For purposes of this volume the major elements constituting metabolites are considered to be the following six: hydrogen, carbon, nitrogen, oxygen, sulfur, and phosphorus. These are the fundamental elements of the various proteins, which form the framework of all living cells. Useful tracer isotopes exist for all six, and at least one isotope has already been applied for each. It may be noted that all these elements play a role in mineral metabolism, but this is of secondary importance to their organic function.

Since the pioneer work of Schoenheimer who initiated the use of isotopes in the study of intermediary organic metabolism slightly more than a decade ago, a very large literature has grown up. The most important generalizing concept that has come out of this work has been that of the dynamic state, or equilibrium of all organic body constituents [Gen112]. Work on intermediate metabolism of organic substances—carbohydrates, fats, proteins, etc.—has shown that all are undergoing a continuous process of breakdown and resynthesis, even seemingly inert substances such as stored body fat.

The application of isotopes has already solved many important problems in the metabolism of organic substances, but the future unquestionably holds a far vaster range of application.

**25.2. Carbon.** Carbon, which forms the “backbone” of all organic compounds, has three useful isotopes for tracer work: the rarer stable species  $C^{13}$  and two unstable species  $C^{11}$  and  $C^{14}$ . In application,  $C^{11}$  is limited to short-term experiments by reason of its short half-life (20.5 min), whereas  $C^{13}$  (stable) and  $C^{14}$  (about 5,000-year half-life) are valuable for long-term experiments—particularly those in intermediary metabolism.  $C^{11}$  is useful for in vivo studies since, as a positron emitter, it has penetrating annihilation gamma rays. It can be prepared in carrier-free form in the cyclotron by the reaction  $B^{10}(d, n)C^{11}$ . Because of excellent cyclotron yields, it can also be used for making simple organic compounds for preliminary studies to be followed later with more exacting studies using  $C^{13}$  or  $C^{14}$ .  $C^{13}$  must of course be measured with the mass spectrometer;  $C^{14}$ , which emits weak beta particles and no gamma rays, must be counted under fairly exacting conditions, *i.e.*, with a thin mica-window counter and exact geometry or,



in the case of low activities, in a gas counter or ionization chamber. It has a definite advantage over  $C^{13}$  in providing a greater sensitivity of measurement. It can be prepared in high specific activity in the nuclear pile by the reaction  $N^{14}(n, p)C^{14}$ , but in practical operation a considerable amount of stable carbon contaminates this process.

The most significant work with  $C^{11}$  has been studies on carbon dioxide utilization by animals and plants [C29,125,169,174] and on carbon monoxide metabolism [C147]. In the latter case it has been shown that carbon monoxide is not oxidized, at least over a 2-hr period in the human body, into carbon dioxide.

With  $C^{13}$  a rather large series of experiments have been performed dealing with the intermediary metabolism of carbohydrates, fats, and, to a lesser degree, amino acids. A recent review by Vennesland [C156] gives an excellent account up through 1947. Much information has been derived on the role of organic precursors. Among the most important results has been discarding of the idea that certain simple substances are the specific precursors of more complex compounds, *e.g.*, lactic acid as a precursor of glycogen. Rather, they are now regarded as contributors to the general metabolic pool from which the complex substances are formed. In connection with carbon dioxide metabolism, a number of studies have established a revolutionary fact, namely, that carbon dioxide is not merely an end product of organic metabolism in the higher animals, but actually an essential compound in certain organic metabolic reactions, although admittedly these do not compare in complexity with bacterial chemo- and photosynthesis or plant photosynthesis.

$C^{14}$ , having only recently become available in appreciable quantities from the nuclear-pile reactor, is just beginning to have widespread application. It seems destined to become the isotope of choice as a tracer in most future research in intermediary metabolism of proteins, carbohydrates, fats, vitamins, organic pharmaceuticals, etc. In fact, it shows promise of becoming the most important single radioisotope available for tracer work. The most important work with  $C^{14}$  reported up to the end of 1947 is probably the demonstration that radioactively labeled carbonate may be laid down in rat bone for at least several weeks without showing any change in concentration; this suggests great caution in the application of this isotope to humans [C24]. Other important studies have been on the distribution in the rat of  $C^{14}$ -labeled amino acids [C69,173, etc]. (Since this volume was submitted to press, an important monograph by Calvin *et al.* [C37a] on the properties and use of the carbon isotopes has appeared.)

**25.3. Hydrogen.** Hydrogen, which with carbon is found in all organic compounds and which with oxygen comprises, in the form of water, the greater part of living organisms, has two useful tracer isotopes: the rare

stable species deuterium,  $H^2$  (or  $D$ ), and the radioactive species tritium,  $H^3$  (or  $T$ ). Deuterium was the earliest stable tracer used and has had wide application. Tritium has so far had very limited use partly because of the technical difficulties inherent in measuring its very weak beta rays, which can be detected only in a gas counter, and partly because of its limited supply until very recently. Accurate gas counters have now been developed, and ample supplies are now available from the nuclear-pile reactors by the reaction  $Li^6(n, \alpha)H^3$ . This isotope should therefore have increasing application, particularly in view of the fact that a considerable increase in sensitivity of measurement is possible with compounds labeled with tritium over that with compounds labeled with deuterium. Furthermore, its long half-life (about 12 years) makes it available for complex syntheses and long-term experiments.

Deuterium has been used to label a wide variety of organic compounds. These are summarized in a recent text by Kamen (Gen80). Whenever deuterium may be introduced in a relatively stable position in an organic molecule, *i.e.*, attached to a carbon atom where it is generally stable bound, it becomes essentially an auxiliary label for carbon. The first experiments with isotopically labeled compounds were with deuterium-labeled fats [Gen125]. Previous to this work it had been thought that depot or storage fat was a biological energy store and was outside the general metabolism, becoming active only in times of need. However with the deuterium label the surprising fact was soon shown that these fats were in a state of fairly rapid turnover and, furthermore, that fatty acids or fats of one type could be converted into fats in which the acid moiety was changed by deletions or additions to the carbon chain or by desaturation. It has been shown on the other hand that such dietarily essential fatty acids as linoleic acid are not formed from other fatty acids.

Since the initial work with fats, there has grown up an extensive literature on a number of classes of organic compounds labeled with deuterium at stable positions.

Deuterium has also been used, in the form of heavy water, in the study of body-water content (by the isotope-dilution method) [D152,153,195], body-water turnover in fish [D150,151] and man [D152,153], and in a number of other problems in the movement of water in the vertebrate body [D31,72,73, etc.].

Tritium has had very limited application. The only work reported up to the end of 1947 has been four papers: one on photosynthesis [T3], one on the determination of total body water [T4] in a manner analogous to that in which deuterium has been used, and two [T1,2] on the use of tritium in showing the conversion of phenylalanine to adrenalin.

The use of deuterium and tritium as tracers, especially as auxiliary tracers

of carbon, has far from been exhausted, and particularly important future applications should come with these isotopes used in doubly or multiply labeled compounds.

**25.4. Oxygen.** Oxygen has two rarer stable isotopes  $O^{17}$  and  $O^{18}$  that offer considerable potentialities for biological application. So far, however, only  $O^{18}$  has been used in tracer work, and for very limited purposes. The unstable species of longest half-life is  $O^{15}$  (126 sec); this has not been employed for tracer work and does not offer much promise.

Although there have been several chemical tracer studies with  $O^{18}$ , the only biological tracer experiments so far reported are a study on photosynthesis [O9] in which it was shown that the oxygen released in this vital process is derived from water and not from carbon dioxide, a brief investigation of oxygen isotope exchange in animal respiration [O4], and a study on sulfate excretion in the rat [O1]. Other biological studies with the oxygen isotopes have been limited to analyses of their relative abundance in the oxygen evolved in photosynthesis [O6-8] and in bacterial metabolism [O5]. In a recent review on  $O^{18}$  as a tracer isotope [O3] the potentialities of this isotope are discussed.

There remains for the future, therefore, the vast field of intermediary organic metabolism in which tracer work with oxygen isotopes may be expected to reveal much fundamental information.

**25.5. Nitrogen.** Nitrogen, which plays an essential role in the peptide linkage of proteins, as well in the molecular configurations of nucleic acids, many vitamins, and other fundamental components of living systems, has two biologically useful isotopes: a short-lived unstable species  $N^{13}$  (10.13 min half-life) and a rarer stable species  $N^{15}$ .

$N^{13}$  may be made in the cyclotron by the reaction  $C^{12}(d, n)N^{13}$ , but it is not a promising agent despite excellent cyclotron yields. It has had very limited biological application as a tracer. The only studies so far reported are ones supposedly demonstrating the fixation of nitrogen by a nonleguminous plant [N54] (however, subsequent work with  $N^{15}$  [N17] has tended to disprove this), and those of Jones [N35], who has employed  $N^{13}$  in respiratory gas-exchange studies.

$N^{15}$  on the other hand has been extensively used as a tracer, particularly in the study of the intermediary metabolism of amino acids and proteins. Vennesland's recent review [N86] covers literature through 1947 in some detail.

An observation of particular interest has been the fact that proteins must be highly labile molecules in which the peptide bonds are undergoing continual rupture and reclosure with freeing and exchange of amino groups. A further important observation has been that, in so far as almost all the essential amino acids are concerned, it is the carbon chain or ring rather

than the intact molecule that cannot be synthesized. An exception is l-lysine, which is required in the intact state [N48]. A process of de- and reamination of almost all acids goes on continuously in biological systems; it has further been found that, when  $N^{15}$ -labeled ammonia is administered to organisms ranging from viruses to higher plants and animals, it becomes widely distributed in the constituent proteins of the organism. In regard to nitrogenous waste products,  $N^{15}$  has been particularly useful in giving a clear-cut answer to the source of creatine and creatinine [N10]. The sources of creatine are glycine, the amidine group of arginine, and the methyl group of methionine. In the mammal all creatinine is derived directly from body creatine, and the reaction is irreversible *in vivo* [N12].

The use of  $N^{15}$  for labeling amino acids, especially coupled with  $C^{13}$  or  $C^{14}$  in the carbon chains, will unquestionably reveal a vast amount of as yet unknown facts of protein metabolism. It is through such studies that we should derive some degree of fundamental understanding of the complexities of protoplasm and genetic continuity.

**25.6. Sulfur.** For sulfur, a component of many proteins and certain nitrilites essential for many organisms, there have been two isotopes applied to biological problems: a rare stable species  $S^{34}$  and an unstable species  $S^{35}$  (87.1-day half-life). There are two other rare stable species that may conceivably find tracer application,  $S^{33}$  and  $S^{36}$ .

$S^{34}$  has had very limited application, so far having been applied biologically only to the study of the conversion of labeled methionine to cystine *in vivo* [S6].

$S^{35}$  has been available in relatively limited quantities until recently. The nuclear-pile reaction  $Cl^{35}(n, p)S^{35}$  is superior in yield to the several cyclotron reactions. Nevertheless,  $S^{35}$  has been applied to a number of studies on sulfur metabolism, particularly the intermediary metabolism of sulfur-containing amino acids and proteins [S21-26] and the excretion of sulfur wastes as sulfate [S7,25]. It has been used to demonstrate that inorganic sulfate is not reduced and incorporated into the body protein [S25] and that inorganic sulfide is largely oxidized to sulfate although small amounts may be incorporated into protein [S7,8,19]. Preliminary studies have also been carried out with labeled thiamin, which is in a state of dynamic equilibrium just as are the major organic metabolites [S3].

**25.7. Phosphorus.** Phosphorus, a constituent of bone, of many organic energy-storing and -transferring systems, and of nucleoprotein, which is basic to all life as we know it, has but one stable isotope,  $P^{31}$ , and for tracer work but one biologically useful form, the radioactive species  $P^{32}$  (14.3-day half-life). There are more publications dealing with this isotope than with any other species, stable or radioactive.  $P^{32}$  may be produced in excellent

yields from the cyclotron by the reaction  $P^{31}(d, p)P^{32}$  and in the uranium pile by the reactions  $P^{31}(n, \gamma)P^{32}$  and  $S^{32}(n, p)P^{32}$ .

The mineral metabolism of phosphorus has been investigated by a number of workers. It was early found that phosphorus rapidly appears in the bone and that there was a slow but definite turnover of bone phosphates [P77,78]. The excretion of phosphorus was shown to take place through both urine and feces [P196], although less than 10 per cent escaped by the latter route. Other studies have demonstrated the rates at which the inorganic plasma phosphate becomes incorporated into the organic metabolic pool [P64,194, etc.] and the permeability of erythrocytes [P102,167,189] and other cellular membranes to the phosphate ion [P51,207].

The major tracer studies with  $P^{32}$  in intermediary metabolism have been on the formation and fate of two important classes of organic compounds: the phospholipids [see Chaikoff and Zilversmit's review, P66] and the nucleoproteins [see Hevesy's review, P188]. It has been demonstrated that the main site of phospholipid production for systemic use is the liver, although the kidney, intestinal mucosa, brain, and muscle synthesize their own phospholipid independently of the liver. Considerable work has been done on the turnover of ribose- and desoxyribosenucleoprotein in various tissues. When labeled inorganic phosphate is administered to the mammal, it is distributed in the nucleic acid moiety of the nucleoprotein of all tissues. Furthermore, this distribution correlates with the known metabolic and growth characteristics of the various tissues. Desoxyribosenucleic acid, being a nuclear constituent, turns over at a much greater rate in such rapidly growing tissues as the spleen, growing liver, and neoplasms, than in the adult liver, the cells of which, while metabolically very active, are not undergoing many mitoses. Ribosenucleic acid on the other hand, being largely a cytoplasmic constituent, turns over in the liver as much as thirty-three times as fast as desoxyribosenucleic acid; it also has a high turnover rate in the spleen and intestinal mucosa. Such facts correlate well with the known importance of those organs in protein metabolism and with the hypothesis that the ribosenucleotides play a role in the synthesis of proteins.

$P^{32}$  has also found an important role in the treatment of certain blood dyscrasias (see Chap. 29).

Despite the wide and intensive application of this isotope, its potentialities in the study of vital processes have been by no means exhausted.

## CHAPTER 26

### ELEMENTS CONSTITUTING MAJOR MINERAL METABOLITES

ELLSWORTH C. DOUGHERTY

**26.1. Introduction.** For purposes of this volume the major elements constituting mineral metabolites have been selected on the basis of their role in the metabolism of higher animals and accordingly are the following five: sodium, magnesium, chlorine, potassium, and calcium. Hydrogen, carbon, oxygen, and phosphorus are also of major importance in mineral metabolism, but have already been considered in the preceding section. The five listed are the elements that form, along with hydrogen, carbon (as carbonates), oxygen (in all complex ions), and phosphorus (as phosphates), the main essential electrolytes of the protoplasm and extracellular and vascular fluids of higher animals. In addition, magnesium and calcium, along with carbon, oxygen, and phosphorus, are the main mineral constituents of bone—the principal skeletal material of the higher vertebrates. Useful tracer isotopes have been applied in animals for all except magnesium, for which tracer study has been limited to one experiment in plants on photosynthesis.

The study of normal mineral metabolism with isotopic tracers began in 1935 with the work of Chievitz and Hevesy on phosphorus uptake and distribution in the rat [P77]. Since that time a considerable body of literature has grown up, particularly in connection with the dynamics of electrolytes in the mammal and with the metabolism of bone. It has recently been thoroughly reviewed by Hevesy [Gen76].

**26.2. Sodium.** Sodium, like phosphorus, has but one stable isotope, and consequently its tracers are radioactive. Two useful species exist:  $\text{Na}^{24}$ , the short-lived form (14.8-hr half-life) produced in the cyclotron by the reaction  $\text{Na}^{23}(\text{d}, \text{p})\text{Na}^{24}$  and in the nuclear pile by the reaction  $\text{Na}^{23}(\text{n}, \gamma)\text{Na}^{24}$ , and  $\text{Na}^{22}$ , the long-lived form (3-year half-life), produced in the cyclotron by the reaction  $\text{Mg}^{24}(\text{d}, \alpha)\text{Na}^{22}$ . Both  $\text{Na}^{24}$  and  $\text{Na}^{22}$  can be used for in vivo studies since penetrating nuclear gamma rays attend the decay of both. Inasmuch as sodium forms the most important extracellular cation in vertebrates, study of its electrolyte dynamics is of great significance. It is also of minor importance in bone.

Studies with  $\text{Na}^{24}$  have been directed toward the determination of the time and mode of normal uptake [Na10,15,31,36,37, etc.], distribution [Na17,33,51, etc.], and excretion [Na31,39,62,67, etc.] of sodium in the mam-

mal. The time of equilibration of sodium injected in man has been found with  $\text{Na}^{24}$  to be between 9 and 12 hr [Na51]. Initially the spread is rapid—for 2 or 3 hr—to a volume of fluid representing about one-fourth of the body weight. It has been suggested that this represents the extracellular fluid volume.

$\text{Na}^{24}$  has had wide application in studies of vascular and cellular permeability. It has been possible to demonstrate that the turnover between the blood sodium and that of the intestinal lumen is very large [Na85,86], being equal in the dog to the total plasma sodium in about 83 min. In connection with the passage of sodium across the placental barrier in several species, including man, it has been found that the transfer per unit weight of placenta increases greatly as gestation proceeds [Na18-24,27-30]. The penetration of  $\text{Na}^+$  into erythrocytes has been investigated by a number of workers [Na9,56,90] and a considerable species difference shown to exist in the rate of this process. In general, however, sodium can be demonstrated to diffuse in and out of the red cell.

The role of the mammalian adrenal in sodium metabolism has also been investigated with  $\text{Na}^{24}$  [Na1-5]; and  $\text{Na}^{24}$  has been used in the experimental therapy of human and mouse leukemia (see Chap. 29).

With  $\text{Na}^{22}$  much longer term experiments are of course possible than with  $\text{Na}^{24}$ . So far this isotope has had relatively limited application. It has been used to demonstrate the increase in the sodium space of the rat body under conditions of dietary chloride deficiency [Na11] and to show the great depression of sodium excretion in congestive heart failure in man—as much as  $\frac{1}{3}$  of the normal clearance [Na71].

**26.3. Potassium.** Potassium at the present time has but one useful species for biological tracer work,  $\text{K}^{42}$  (12.4-hr half-life), produced in the cyclotron largely by the reaction  $\text{K}^{41}(\text{d}, \text{p})\text{K}^{42}$  and in the nuclear pile by the reaction  $\text{K}^{41}(\text{n}, \gamma)\text{K}^{42}$ .  $\text{K}^+$  is an almost universally essential intracellular cation; in the mammal it serves as the main cation within the cells in contrast with  $\text{Na}^+$  in the extracellular fluids. In ascending the periodic system potassium is the first element that has a naturally occurring, rare, long-lived radioactive isotope; this species,  $\text{K}^{40}$  (about  $10^9$ -year half-life), has been speculated upon as conferring on potassium some of its physiological properties [K18,36,42,51], but there has been no adequate confirmation of any of these views [K18]. Some evidence exists, however, that biological systems may deal with the isotopes of potassium in a slightly different manner [K16,35].

The half-life of  $\text{K}^{42}$  limits it to short-term experiments. Nevertheless, a number of studies of potassium uptake [K18,24] and distribution [K15,17,21,28,52] have been carried out. In mammals injected with labeled  $\text{K}^+$ , the ion penetrates rapidly into most tissues of the body, the testes, brain, and erythrocytes being slowest to exchange intracellular with plasma potassium

[K17,52]. In tissues with rapid penetration—liver, heart, kidney, etc.—the total potassium has a relative activity higher than the simultaneous plasma value for the first 1 to 2 hr after administration, particularly after intraperitoneal injection.

The penetration of  $K^+$  into erythrocytes has been investigated by several workers [K11–13,40,45,46,50,59]. As with  $Na^+$  a considerable species difference in permeability and exchangeability has been shown.

The physiology of the adrenal cortex has also been studied with  $K^{42}$  [K1–5].

Long-term tracer studies of potassium metabolism may become possible now that the rarer naturally occurring species,  $K^{40}$  (radioactive) and  $K^{41}$  (stable), can be concentrated in a practical manner, or if an artificial species of long half-life is discovered.

**26.4. Calcium.** Calcium, the most abundant positively ionized mineral element of the mammalian body, is largely found in bone, but the small amount occurring in the plasma and inside cells is of great physiological significance—in neuromuscular irritability, blood coagulation, cellular permeability, and certain enzymatic systems. In tracer work the one radioactive species so far used,  $Ca^{45}$  (180-day half-life), has had rather limited application because of poor cyclotron yields from the reaction  $Ca^{44}(d, p)Ca^{45}$ . It is now, however, available in much larger quantities from the nuclear pile by the reaction  $Ca^{44}(n, \gamma)Ca^{45}$  and should have greatly increased utilization. The cyclotron reaction producing  $Ca^{45}$  also results in some  $Ca^{41}$  (8.5-day half-life) by  $Ca^{40}(d, p)Ca^{41}$ . However, this latter species decays by K capture and emits only feeble x-rays, which are difficult of detection.  $Ca^{45}$  can presumably be prepared in a carrier-free state in the nuclear-pile reactor by the reaction  $Sc^{46}(n, p)Ca^{45}$ .

Studies with  $Ca^{45}$  have so far been limited to a few investigations, primarily on the uptake of calcium by calcified tissues.  $Ca^{45}$  is stored almost entirely in bone [Ca2,7,9] with traces appearing throughout the soft tissue. Vitamin D promotes the absorption of calcium from the digestive tract and the mineralization of the bones of rachitic rats [Ca4].

Much work on bone metabolism has been done by substituting radioactive strontium for calcium by reason of the similar metabolic behavior of these two elements. For citations to this work see Sec. 28.2 *h*.

**26.5. Magnesium.** Despite the important biological role of magnesium, as an essential element for all known forms of life, it has scarcely been studied by tracer isotopes. The radioactive species of longest half-life  $Mg^{24}$ , producible in the cyclotron by the reaction  $Mg^{26}(d, p)Mg^{27}$ , has a half-life of only 10.2 min. It has been used in but one biological experiment [Mg1] in which its incorporation into chlorophyll by an alga and by barley was shown. By the time chlorophylls *a* and *b* could be separated, however, the activity



of the  $\text{Mg}^{27}$  was too weak for satisfactory measurement, and therefore no interconversion of the two types of photosynthetic pigments could be demonstrated using this magnesium label.

Now that the rarer stable species  $\text{Mg}^{25}$  and  $\text{Mg}^{26}$  have been prepared in enriched form, it should be possible to investigate not only the role of magnesium in photosynthesis, but also its other known vital functions—in enzyme systems, bone metabolism, cellular permeability, etc.

**26.6. Chlorine.** Chlorine, essential as an anion in the higher animals and as a micronutrient requirement for some plants, has three potentially useful radioactive species: the short-lived forms  $\text{Cl}^{34}$  (33 min half-life) and  $\text{Cl}^{38}$  (27-min half-life) and the long-lived form  $\text{Cl}^{36}$  (10<sup>6</sup>-year half-life). So far only  $\text{Cl}^{38}$  has been used in biological experiments; it is made in the cyclotron by the reactions  $\text{Cl}^{37}(\text{d}, \text{p})\text{Cl}^{38}$ ,  $\text{Cl}^{37}(\text{n}, \gamma)\text{Cl}^{38}$ , and  $\text{K}^{41}(\text{n}, \alpha)\text{Cl}^{38}$ .  $\text{Cl}^{34}$  can be produced in the cyclotron by two reactions:  $\text{P}^{31}(\alpha, \text{n})\text{Cl}^{34}$  and  $\text{S}^{33}(\text{d}, \text{n})\text{Cl}^{34}$ .  $\text{Cl}^{36}$ , which can be produced in significant amounts only in the nuclear pile, by the reaction  $\text{Cl}^{35}(\text{n}, \gamma)\text{Cl}^{36}$ , has only recently become available for tracer work.

The short half-life of  $\text{Cl}^{38}$  has limited tracer experiments thus far reported for chlorine to short-term studies. When introduced as labeled lithium chloride,  $\text{Cl}^{38}$  rapidly finds its way into the chloride space of such organs and tissues as the kidney, liver, muscles, cartilage, and tendons, but not into that of the testes and pyloric mucosa [Cl11].  $\text{Cl}^-$  has also been found to penetrate rapidly into erythrocytes [Cl15]. It appears in normal gastric juice very quickly, within 1 to 2 min after intravenous injection into the man or dog [Cl2], but more slowly in the case of achlorhydria [Cl3].

When  $\text{Cl}^{38}$  is widely available, tracer work with chlorine can be much expanded.

## CHAPTER 27

# TRACE ELEMENTS KNOWN TO BE ESSENTIAL IN ANIMALS AND PLANTS

ELLSWORTH C. DOUGHERTY

**27.1. Introduction.** The elements so far considered have major metabolic roles in some or all living systems. In addition to these macronutrient constituents of protoplasm, there are a number of elements that have been recognized as being essential in "trace" amounts in the metabolism of certain organisms at least. Certain of these micronutrient elements, at least iron and probably also manganese and zinc, are universally required for living systems. A number of others are known to be essential for certain forms of life but not for others.

The elements accepted as essential trace nutrients for purposes of this volume are, in order of increasing atomic weight, boron, aluminum, silicon, vanadium, manganese, iron, cobalt, copper, zinc, molybdenum, and iodine. They are arranged somewhat arbitrarily in the following subsections according to their importance in mammalian nutrition; the last five elements (molybdenum, boron, aluminum, silicon, and vanadium) are known to be essential either to plants or to certain of the lower animals, but not to mammals. Comar [Gen28] has recently reviewed in detail trace elements in mammalian nutrition.

It will likely prove very difficult to carry out satisfactory tracer studies on trace elements for which no sufficiently long-lived isotope is available to permit more than very short experiments. Silicon and particularly boron and aluminum are in this category. The use of separated stable isotopes in the case of silicon and boron does not offer much advantage theoretically over the natural element.

**27.2. Iron.** Iron, which is regarded as a universally essential trace element in the oxidative enzyme systems of cells, exists in two radioactive forms that have found tracer application:  $\text{Fe}^{55}$  (about 4-year half-life) and  $\text{Fe}^{59}$  (47-day half-life). A mixture of these isotopes can be produced in the cyclotron by the deuteron bombardment of iron:  $\text{Fe}^{54}(\text{d}, \text{p})\text{Fe}^{55}$  and  $\text{Fe}^{58}(\text{d}, \text{p})\text{Fe}^{59}$ ; in addition  $\text{Fe}^{55}$  can be made by the reactions  $\text{Mn}^{55}(\text{p}, \text{n})\text{Fe}^{55}$  or  $\text{Mn}^{55}(\text{d}, 2\text{n})\text{Fe}^{55}$ , and  $\text{Fe}^{59}$  by the reaction  $\text{Co}^{59}(\text{n}, \text{p})\text{Fe}^{59}$ . In the nuclear-pile reactor a mixture of both isotopes can be produced by the bombardment of iron  $\text{Fe}^{54}(\text{n}, \gamma)\text{Fe}^{55}$  and  $\text{Fe}^{58}(\text{n}, \gamma)\text{Fe}^{59}$ . Counters sensitive to  $\text{Fe}^{55}$  and

$\text{Fe}^{59}$  individually are known and make possible the use of these two isotopes in multiple-labeling experiments.

$\text{Fe}^{59}$  has been the more widely used isotope. It has been employed largely in the study of iron metabolism and in the determination of the red-cell survival time and the total red-cell volume in the mammal, especially man. This use has recently been reviewed by Hahn [Fe32]. A number of important new facts about iron metabolism have been demonstrated with  $\text{Fe}^{59}$ . Particularly striking has been clear-cut proof of the ability of the mammal to conserve iron supplies by limiting uptake and excretion [Fe24,37,40, etc.]. The uptake of iron is controlled by the presence of a protein that is capable of combining stoichiometrically with 23 per cent of its weight of iron [Fe40,45]; this is ferritin (or, in its iron-free state, apoferritin). Ferrous iron is better absorbed than ferric by man [Fe46,47,58]. The loss of iron from the body is largely through wear and tear rather than any of the usual routes of excretion.

The most important practical result of studies on the survival of red cells has been demonstration of the best method of in vitro storage of blood to be used for transfusions [Fe15-17,19,67-69]; this involves refrigeration, the addition of dextrose to the citrate anticoagulant, the maintenance of a slightly acid reaction of the diluted plasma or resuspension fluid, and optimal dilution. In certain acid-citrate solutions whole blood can be preserved with up to 70 per cent viability for 3 weeks, whereas cells preserved by the simple addition of citrate are almost entirely broken down within 24 hr in the new host after only 2 weeks of preservation in vitro [Fe65].

The  $\text{Fe}^{59}$ -labeled red cell has been used to follow the distribution of blood [Fe20,21] and to determine the total red-cell volume by the intravenous administration of tagged cells of known activity and the recovery of blood samples after allowing time for mixing to approach completion [Fe18,21,22,29, etc.].

$\text{Fe}^{55}$  has had relatively little application, but because of its availability in high specific activity from the cyclotron by the reaction  $\text{Mn}^{55}(\text{d}, 2\text{n})\text{Fe}^{55}$ , it has been useful for studies on the distribution of tracer amounts. Work with this isotope has shown the rapid turnover of iron in the bone marrow, its storage in major part in the liver, and its rapid turnover in the small intestine [Fe7], as well as its minimal excretion in the urine, bile, and feces [Fe7,24]. An ingenious study with  $\text{Fe}^{55}$  has shown that the parasite of benign tertian malaria (*Plasmodium vivax*) selectively invades young erythrocytes [Fe14].

$\text{Fe}^{56}$  and  $\text{Fe}^{58}$  have been used in double-labeling experiments on red-cell survival [Fe16,17, etc.]. Conceivably the rarer stable species of iron  $\text{Fe}^{54}$ ,  $\text{Fe}^{57}$ , and  $\text{Fe}^{58}$  may find use in multiple-labeling experiments now that they have been prepared in enriched form.

**27.3. Iodine.** Iodine, an essential element in vertebrate metabolism, has a number of useful and potentially useful radioactive isotopes. Three

species have found biological application:  $I^{128}$  (24.99-min half-life) produced in the cyclotron by the reactions  $I^{127}(d, p)I^{128}$  and  $I^{127}(n, \gamma)I^{128}$ ;  $I^{130}$  (12.6-hr half-life) and  $I^{131}$  (8.0-day half-life), produced together in the cyclotron by the reactions  $Te^{130}(d, 2n)I^{130}$  and  $Te^{130}(d, n)I^{131}$ , respectively.  $I^{130}$  and  $I^{131}$  also occur as fission products, the latter in much greater quantities;  $I^{130}$  can be prepared by itself in the cyclotron by the reaction  $Te^{130}(p, n)I^{130}$ ; and  $I^{131}$  can be recovered in carrier-free form from the nuclear pile and subsequent decay reaction  $Te^{130}(n, \gamma)Te^{131} \rightarrow I^{131} + \beta^-$ . In addition there are several other as yet unapplied species of varying half-lives:  $I^{124}$  (4-day),  $I^{125}$  (56-day),  $I^{126}$  (13.0-day),  $I^{133}$  (22-hr), and  $I^{135}$  (6.6-hr). Some of these may find biological application.

Early work with iodine was done with  $I^{128}$  (in some cases produced by bombarding  $I^{127}$  with neutrons from a radium-beryllium mixture). The concentration of trace amounts of iodine in the thyroid gland was demonstrated strikingly by this early work [143,70,72, etc.].

Most recent tracer work has been done with the two isotopes  $I^{130}$  and  $I^{131}$ , which are produced as a mixture in the cyclotron by deuteron bombardment of tellurium. Currently  $I^{131}$  is used almost exclusively. The primary application of the iodine label has, of course, been in the study of thyroid physiology. This has recently been reviewed by Leblond [163].

Before radioactive iodine became available, the entry of iodine into the thyroid gland could not be studied under physiological conditions. It has become possible with the radioactive species to study not only entry but also the incorporation of iodine into the thyroid hormone. When minute doses of iodine are administered to the mammal, a high proportion becomes incorporated in the gland—about 60 per cent in the rat by 24 hr [193] and about 50 per cent in man [19,41,69]. However, when these small doses are given after pretreatment with pharmacological amounts, the uptake of the labeled material is much reduced [144,72]. Initially the inorganic iodide fraction of the thyroid iodine has a higher specific activity than the organically bound. This becomes incorporated into diiodotyrosine and thyroxine. Diiodotyrosine turnover in man is thought to take about 24 hr [169]. For several days the specific activity of the thyroxine iodine is less than that of the inorganic or diiodotyrosine iodine, but, while the diiodotyrosine fraction remains more or less constant, the thyroxine activity gradually rises. This is satisfactorily explained by assuming the conversion of diiodotyrosine into thyroxine rather than the direct iodination of thyronine, the non-iodine-containing analogue of thyroxine [181].

With radioactive iodine it has been possible to study precisely the effects of various extrinsic factors on iodine metabolism in the thyroid, *e.g.*, pituitary [190,91, etc.], thiourea [14], sulfonamides [114,123], goitrogenic substances [115,123], etc.

Radioactive iodine has also provided a therapeutic tool in the treatment of hyperthyroidism (see Chap. 29).

The use of the iodine label is the best illustration of the invaluable role that radioactive isotopes can play in the study of trace-element metabolism.

**27.4. Manganese.** Manganese, which is known to activate a number of enzymes, but of which the biological role is not yet clearly defined, is nevertheless thought to be a probable micronutrient requirement for all living systems. It has been well established as essential to the growth and reproduction of higher animals. At least three useful radioactive species exist:  $\text{Mn}^{52}$  (6.5-day half-life) made in the cyclotron by the reaction  $\text{Cr}^{52}(\text{d}, 2\text{n})\text{Mn}^{52}$ , which also produces small amounts of  $\text{Mn}^{54}$  by the reaction  $\text{Cr}^{54}(\text{d}, 2\text{n})\text{Mn}^{54}$ ;  $\text{Mn}^{54}$  (310-day half-life), made in the cyclotron by the reaction  $\text{Fe}^{56}(\text{d}, \alpha)\text{Mn}^{54}$ , which is accompanied by the reaction  $\text{Fe}^{54}(\text{d}, \alpha)\text{Mn}^{52}$ ; and  $\text{Mn}^{56}$  available from the cyclotron by the reaction  $\text{Mn}^{55}(\text{d}, \text{p})\text{Mn}^{56}$ .

Tracer experiments have been reported with all three of the isotopes mentioned.  $\text{Mn}^{52}$  (admixed with small amounts of  $\text{Mn}^{54}$ ) has been applied to the study of the distribution of colloidal manganese oxide injected intravenously for therapeutic purposes in man (see Chap. 29).  $\text{Mn}^{54}$  (admixed with small amounts of  $\text{Mn}^{52}$ ) has had wider use, having been employed to show that tracer amounts of manganese are rapidly excreted in the rat almost entirely in the feces [ $\text{Mn}^{54}$  (in the latter reference  $\text{Mn}^{54}$  is erroneously referred to as  $\text{Mn}^{56}$ )] and that, even in the case of perosis in the chicken produced by manganese deficiency, the element is not deposited to a greater than normal extent in the bone, although it accumulates in the liver in greater amounts than in normal animals [ $\text{Mn}^{54}$ ].  $\text{Mn}^{56}$  has also been used for the study of manganese distribution in the mammal [ $\text{Mn}^{56}$ ].

The use of the tracers of manganese has been very limited, and much further work may be anticipated.

**27.5. Copper.** Copper, known to be an essential micronutrient requirement at least of higher animals and functioning in a number of protein and enzyme complexes, has two potentially useful radioactive species for biological experimentation:  $\text{Cu}^{61}$  (3.4-hr half-life) and  $\text{Cu}^{64}$  (12.8-hr half-life) made as a mixture by the cyclotron reactions  $\text{Ni}^{60}(\text{d}, \text{n})\text{Cu}^{61}$ ,  $\text{Ni}^{61}(\text{d}, 2\text{n})\text{Cu}^{61}$ , and  $\text{Ni}^{64}(\text{d}, 2\text{n})\text{Cu}^{64}$ . In addition,  $\text{Cu}^{64}$  can be made in the uranium pile by the reaction  $\text{Cu}^{63}(\text{n}, \gamma)\text{Cu}^{64}$ . The two stable species, especially the rarer one  $\text{Cu}^{65}$ , may also find tracer application, particularly in view of the relatively short half-life of  $\text{Cu}^{64}$ .

So far the few biological studies published have been done with  $\text{Cu}^{64}$ , any associated  $\text{Cu}^{61}$  having decayed away or having been ignored. Studies on the uptake and elimination of tracer amounts of copper fed to rats show that, of that absorbed, most goes to the liver in absolute amount, but that it concentrates relatively more in the kidneys than in other organs, although little

is eliminated in the urine [Cu4]. Following ingestion it appears rapidly in the plasma [Cu7]; the uptake is relatively greater in animals having hemato-poietic activity increased over normal [Cu4,7]. Most of the copper present in the plasma is bound in some way to protein.

The existence of a longer lived radioisotope, presumed to be  $\text{Cu}^{67}$  (56-hr half-life), promises considerable extension in tracer work with copper provided that this species can be produced in sufficient yield.

**27.6. Zinc.** Zinc, like manganese, is important in enzymatic functions and may well be a universal micronutrient requirement of living systems. It is known to be essential in the metabolism of the vertebrate. It occurs in several stable and radioactive species of which only two have had biological application:  $\text{Zn}^{65}$  (250-day half-life), produced in the cyclotron by the reaction  $\text{Zn}^{64}(\text{n}, \gamma)\text{Zn}^{65}$  and  $\text{Cu}^{65}(\text{d}, 2\text{n})\text{Zn}^{65}$  and in the uranium pile by the reaction  $\text{Zn}^{64}(\text{n}, \gamma)\text{Zn}^{65}$ ; and  $\text{Zn}^{63}$  (38-min half-life), produced in the cyclotron by the reactions  $\text{Cu}^{63}(\text{d}, 2\text{n})\text{Zn}^{63}$  and  $\text{Cu}^{63}(\text{p}, \text{n})\text{Zn}^{63}$ . Other radioisotopes exist of short half-life:  $\text{Zn}^{69}$  (13.8-hr),  $\text{Zn}^{69}$  (57-min—isomer of preceding), and  $\text{Zn}^{72}$  (49-min). These, with  $\text{Zn}^{63}$ , offer possible alternatives if it is desired to avoid the long half-life of  $\text{Zn}^{65}$ .

So far the biological experiments with  $\text{Zn}^{65}$  have been limited to a few studies on the fate of zinc administered by various routes [Zn1,6,7]. Like manganese, but unlike cobalt, zinc is excreted largely by way of the gastrointestinal tract, with relatively little in the urine. It accumulates in the liver from which it is rapidly lost in the bile; it also concentrates to varying degrees in the pancreas, kidney, and spleen.

$\text{Zn}^{63}$  has been used [Zn2-4] to obtain localized irradiation of tissues injected with labeled colloidal zinc.

Further fundamental advances in the understanding of zinc metabolism quite evidently depend on further application of zinc tracers.

**27.7. Cobalt.** Cobalt appears according to recent evidence to be an essential trace element in the nutrition of higher animals. There are a number of radioactive isotopes of which four are formed by the deuteron bombardment of iron:  $\text{Co}^{55}$  (18.0 hr) by  $\text{Fe}^{54}(\text{d}, \text{n})\text{Co}^{55}$ ;  $\text{Co}^{56}$  (80 days) by  $\text{Fe}^{56}(\text{d}, 2\text{n})\text{Co}^{56}$ ;  $\text{Co}^{57}$  (270 days) by  $\text{Fe}^{56}(\text{d}, \text{n})\text{Co}^{57}$ ; and  $\text{Co}^{58}$  (72 days) by  $\text{Fe}^{57}(\text{d}, \text{n})\text{Co}^{58}$ . From the nuclear-pile reactor,  $\text{Co}^{60}$  (5.3-year half-life) is available by the reaction  $\text{Co}^{59}(\text{n}, \gamma)\text{Co}^{60}$ .

With one exception the few studies reported have been carried out with mixtures of  $\text{Co}^{56}$ ,  $\text{Co}^{57}$ , and  $\text{Co}^{58}$ , of which the first and last make up the more important component;  $\text{Co}^{55}$  decays away in a few days, and consequently targets are allowed to age so that this species can be disregarded. The chief route of excretion of cobalt is the urine, in contrast to manganese and zinc [Co5,7,9]. Ingested cobalt is only partly absorbed, and the bile

is an important but not the sole pathway for the passage of cobalt from the body into the intestinal tract [Co7,9].

Recently the long-lived species  $\text{Co}^{60}$  has come into use. It has been employed in studies on cobalt distribution, particularly, in the bone marrow [Co8].

**27.8. Molybdenum.** For molybdenum, an element required by certain higher plants, there are two known radioactive species of convenient half-life:  $\text{Mo}^{99}$  (67-hr half-life), which suffers, however, by being the parent of radioactive  $\text{Te}^{99}$ ; and  $\text{Mo}^{93}$  (6.7-hr half-life). In addition there are a number of stable species that might prove of value.

$\text{Mo}^{99}$  has recently been used in preliminary tracer work in animals [Mo1] and plants [Mo2].  $\text{Mo}^{93}$  has also been used in plant studies [Mo2].

**27.9. Boron.** Boron, which is known to be an essential micronutrient of higher plants and some microorganisms, has not been studied by tracer methods. The only possible tracer isotope known is the stable species  $\text{B}^{10}$ , which constitutes 18.4 per cent of the element. This species is now available in nearly pure form.

Work has, however, been done with boron in the study of the effect of the selective ionization produced by bombarding this element concentrated in tissues [B1-3] with slow neutrons. These studies have suggested that the splitting of  $\text{B}^{10}$  under such conditions may possibly find therapeutic application.

**27.10. Aluminum.** Aluminum, which is known to be essential to certain plants and protozoans, has also not been studied by tracer techniques. The only available species would seem to be  $\text{Al}^{28}$  and  $\text{Al}^{29}$  with half-lives of only 2.4 and 6.7 min, respectively. At best these offer very limited potentialities.

**27.11. Silicon.** Silicon, which is also known to be essential in the nutrition of certain plants and protozoa and which may also be a necessary micronutrient in higher animals, is another element not yet studied by biological tracing methods. Possibly useful species are the two rarer stable species  $\text{Si}^{29}$  and  $\text{Si}^{30}$  and the radioactive species  $\text{Si}^{31}$  (170-min half-life).

**27.12. Vanadium.** Vanadium is an essential part of a metalloporphyrin respiratory pigment in certain lower chordates. Its metabolism could readily be studied with the radioactive isotope  $\text{V}^{48}$  (16-day half-life) and possibly with  $\text{V}^{49}$  (600-day half-life).

## CHAPTER 28

### ELEMENTS NOT KNOWN TO BE ESSENTIAL TO LIFE

ELLSWORTH C. DOUGHERTY

**28.1. Introduction.** Some 22 of the 96 known elements have been considered so far. The remaining 74 are not known to be essential to living systems. However, as already noted in Chap. 24, there are certain elements for which a vital role has been suggested on evidence of varying strength. For purposes of this volume these elements (some 11) are considered in Sec. 28.2.

The remaining elements are grouped more or less arbitrarily into (1) elements of common pharmacological and toxicological importance, (2) the noble gases, (3) elements of the lanthanide rare-earth series and others in fission, (4) the actinide rare-earth series, and (5) other elements.

For the main nuclear reactions producing the isotopes considered in the following subsections, Sec. 7.12 should be consulted.

**28.2. Possible Micronutrient Elements.** Eleven elements are treated here as having been regarded as possible trace requirements for animals or plants. This does not complete the list of elements for which such a role has been suggested, but it does cover the principal ones. These elements are lithium, fluorine, chromium, nickel, gallium, bromine, rubidium, strontium, columbium, tin, and tungsten

*a. Lithium.* No tracer work has been reported for lithium, which has been found widely present in trace amounts in animals. It has no known radioactive isotope of half-life greater than 0.88 sec; thus the rarer stable species  $\text{Li}^6$  will presumably have to serve for purposes where changes in systems in dynamic lithium equilibrium are to be studied. Some studies have been made with natural lithium on the effect of bombarding lithium in tissues with slow neutrons [Li1-2]; like  $\text{B}^{10}$ , the bombarded  $\text{Li}^6$  nucleus splits with the release of high specific ionization. These studies have given results similar to those obtained with boron.

*b. Fluorine.* The effect of fluorine in making teeth more resistant to caries has led to its being postulated as an essential micronutrient in vertebrates. A few studies have been carried out with  $\text{F}^{18}$  (112-min half-life) on the uptake of fluorine in bone and teeth [F1-3], both in vivo and in vitro. The secretion of fluorine in the submaxillary-gland saliva of the cat has also been shown [F1].



*c. Chromium.* No tracer work has been carried out for chromium, which has been suggested as a possible trace requirement in certain plants and animals. The radioactive species  $\text{Cr}^{51}$  (26.5-day half-life) is available for such studies.

*d. Nickel.* Nickel has been found concentrated in the mammalian pancreas and in the bodies of several invertebrates and may be involved in enzyme systems. Its metabolism has not yet been subjected to tracer studies, although the radioactive isotopes  $\text{Ni}^{57}$  (36-hr half-life),  $\text{Ni}^{59}$  (12-year half-life), and  $\text{Ni}^{66}$  (56-hr half-life) are available and should prove useful.

*e. Gallium.* There is strong evidence for the essential role of gallium in certain plants. It has not yet been studied with tracer methods, however. Several potentially useful radioactive species exist, the most promising of these for tracer studies being  $\text{Ga}^{67}$  (83-hr half-life).

*f. Bromine.* Bromine is present in small amounts as an electrolyte in the vertebrate body, primarily extracellularly like chlorine. Although an essential role has been postulated, definitive evidence is lacking.

Tracer studies with the radioactive species  $\text{Br}^{82}$  (34-hr half-life) have been fairly extensive. The bromide ion has been found to behave in general distribution very much like  $\text{Na}^+[\text{Br}6]$ . It is completely distributed in the dog body within a period of 1 to 2 hr.

$\text{Br}^{82}$  has also been used to label diazo dyes in studying the escape of colloids from the blood stream  $[\text{Br}3]$  and the concentration and detection of dyes in abscesses  $[\text{Br}10-12,17]$ . Certain dyes become bound to plasma proteins and, when labeled with  $\text{Br}^{82}$ , have been used in dogs to show the great increase in capillary permeability to colloids after burns  $[\text{Br}3]$ .

The significant selective uptake of bromine by the thyroid has also been shown with  $\text{Br}^{82}$   $[\text{Br}13]$ .

*g. Rubidium.* It has been claimed that rubidium may actually replace potassium in the nutrition of certain bacteria. The element is present in trace amounts as an ion in the vertebrate body fluids. Evidence for an essential role is, however, lacking in either case.

There have been a few studies on the behavior of ionic rubidium in biological systems with the use of the radioactive isotope  $\text{Rb}^{86}$  (19.5-day half-life). In the dog it closely resembles potassium in distribution after intravenous injection  $[\text{Rb}1]$ . The uptake of the element and its concentration from very dilute solutions of  $\text{Rb}^+$  by the roots of plants has also been demonstrated  $[\text{Rb}3]$ .

*h. Strontium.* Strontium has been of considerable interest because of the similarity of its fate in vertebrate metabolism to that of calcium. Because of the poor cyclotron yields of  $\text{Ca}^{45}$ , considerable study on bone physiology has been carried out using the radioactive isotope  $\text{Sr}^{85}$  (65-day half-life) in place of a calcium isotope. Despite its well-known metabolic behavior in

the vertebrate, the essential nature of strontium as a trace element has not been established.

Studies with  $\text{Sr}^{85}$  have shown that, when injected, the element rapidly concentrates in the skeleton. In the mouse the radioactivity per unit wet weight of skeleton is about 100 times that of the soft tissues in 24 hr. Of the soft tissues the liver and especially fat show the lowest activity at this time [Sr19,20].

Studies on bone-fracture healing in normal rats and in rats on abnormal vitamin balance have shown that recalcification is less active in vitamin A- and D-deficient and D-hypervitaminotic rats. In normal rats the callus undergoes most active calcification within from 8 to 16 days, normal strength of the broken bone being recovered at 12 to 16 days [Sr4]. In another experiment it was found that hypophysectomized rats treated with growth hormone deposited in a short period essentially the same amount of  $\text{Sr}^{85}$  in the femur and mandible as did untreated hypophysectomized rats. This suggests that mineral deposition in bone is independent of growth hormonal control [Sr17].

For the therapeutic application of  $\text{Sr}^{85}$  to bone malignancy see Chap. 29.

i. *Columbium*. There is suggestive evidence that columbium is an essential trace requirement in certain plants. Tracer work has, however, been very limited and so far carried out with  $\text{Cb}^{95}$  (37-day half-life). It has been used in radioactive colloids that localize in the reticuloendothelial system [Cb1, see Chap. 29] and in one experiment with the tracer-free element [Cb2] in which it was shown that in the rat parenterally administered columbium accumulated about 40 per cent in the bone and was lost therefrom with a half-time of about 100 days. (For further discussion of the uptake in bone of fission elements see Sec. 28.5.)

j. *Tin*. Although tin has been found as a trace element rather widely in animals, no essential physiological role has been demonstrated. Tracer work is yet to be done. There are at least three potentially useful tracer isotopes: the radioactive species  $\text{Sn}^{113}$  (70- to 105-day half-life),  $\text{Sn}^{121}$  (60-hr half-life), and  $\text{Sn}^{123}$  (10-day half-life).

k. *Tungsten*. Tungsten has been thought to be essential as a micro-nutrient of certain plants, but no tracer work has yet been attempted with it. There are available two potentially useful radioactive species:  $\text{W}^{185}$  (77 day half-life) and  $\text{W}^{187}$  (24.1-hr half-life).

**28.3. Elements of Importance in Pharmacology and Toxicology.** The common elements of toxicological and pharmacological importance not already considered are arsenic, silver, antimony, gold, mercury, lead, bismuth, and radium. Tracer studies have been carried out for all of these except silver.

a. *Arsenic*. Arsenic is an important agent in syphilotherapy and the treatment of leukemia. It is also used in antiprotozoal and anthelmintic drugs.

Studies with radioactive arsenic have been limited to the isotope  $\text{As}^{74}$  (16-day half-life). Tracer experiments have demonstrated the uptake of subcutaneously administered arsenites in the body proteins of various mammals, but have not supported the view that arsenic may replace phosphorus in proteins [As7,11]. In concentration per unit weight of tissue arsenic administered as arsenite is found in largest amounts in the kidney and in decreasing amounts in the liver, epidermis, spleen, lung, and muscle [As10]. In experimental filariasis (*Litomosoides carinii* infection in the cotton rat) the concentration in the adult worm is only less than in the kidney and liver [As10].

b. *Silver*. Potentially valuable radioactive species for tracer work with silver are particularly the isotopes  $\text{Ag}^{106}$  (8.2-day half-life),  $\text{Ag}^{110}$  (225-day half-life), and  $\text{Ag}^{111}$  (7.5-day half-life).

c. *Antimony*. Antimony, important in anthelmintic and antiprotozoal agents, has had some tracer application in the form of  $\text{Sb}^{124}$  (60-day half-life). When administered to dogs in the form of tartar emetic, it is found to concentrate most highly in the liver and in decreasing amounts in the thyroid, kidney, cortex, pancreas, intestine, spleen, etc. In dogs with filariasis (the heart worm, *Dirofilaria immitis*)  $\text{Sb}^{124}$  concentrated more highly in the adult worm than in any other tissues except the liver and thyroid [Sb2,3].

d. *Gold*. A single tracer study has been reported for gold with the use of  $\text{Au}^{198}$  (2.7-day half-life). The highest uptake of the element is in the kidney and liver [Au1].

e. *Mercury*. Mercury, an important industrial poison and therapeutic agent, has had but one tracer study—with the radioactive species  $\text{Hg}^{197}$  (23- and 64-hr half-lives). It was used to establish the concentration of mercury vapor in a particular industrial operation suspected of causing chronic mercurialism [Hg1].

f. *Lead*. Lead is an important industrial poison. It was the first element to be studied biologically by the "tracer" technique, namely, that of substituting for a normally stable element one of its radioactive species [Pb7] and thus tracing its fate in a biological system (in this case a plant).

Two lead isotopes, both belonging to naturally radioactive series, have been used in the study of the uptake and fate of lead in organisms:  $\text{Pb}^{210}$  or  $\text{RaD}$  (22-year half-life) and  $\text{Pb}^{212}$  or  $\text{ThB}$  (10.6-hr half-life).  $\text{Pb}^{210}$  belongs to the uranium series;  $\text{Pb}^{212}$ , to the thorium series.

Studies with  $\text{Pb}^{210}$  have included the uptake of lead in erythrocytes [Pb9], the absorption of lead tetraethyl [Pb8], the effect of pectin on the retention

of dietary lead [Pb5], etc. Studies with  $\text{Pb}^{212}$  have been on the uptake of lead by erythrocytes [Pb1,2] and the general distribution of lead in normal and tumor-bearing mice [Pb1,6].

*g. Bismuth.* The use of bismuth as an antisyphilitic and antiprotozoal agent renders tracer studies of considerable importance. So far only two tracer studies for the element [Bi1,2] have appeared, both done with  $\text{Bi}^{210}$  or  $\text{RaE}$  (5-day half-life) of uranium series. Bismuth was found upon parenteral injection to be eliminated in the urine and feces, about twice as much in the former as in the latter, and to concentrate most highly in the kidney and somewhat less in the liver.

*h. Radium.* Radium has presented a toxicological problem particularly in the watchmaking industry in connection with the painting of luminous dials. No attempt has been made here to cover the extensive literature on the toxicology and radiation effects of this element.  $\text{Ra}^{226}$  is the common isotope (1,596-year half-life). A few tracer studies have been carried out using  $\text{Ra}^{224}$  or  $\text{ThX}$  (3.64-day half-life) and  $\text{Ra}^{228}$  or  $\text{MsTh}_1$  (6.7-year half-life).

**28.4. Noble Gases.** The use of radioactive isotopes of three of the heavier gases—argon, krypton, and xenon—has made possible important studies on respiratory gas exchange and the pattern of blood mixing in the mammal [A1,2,K1,5, etc.]. The marked solubility of xenon in fat has been shown with the radioactive isotopes  $\text{Xe}^{127}$  (34-day half-life) [Xe4] and  $\text{Xe}^{133}$  (7-day half-life) [Xe1].

Isotopes of radon— $\text{Rn}^{220}$  or  $\text{Tn}$  (54.5-sec half-life) and  $\text{Rn}^{222}$  (3.825-day half-life)—have been measured or used in studies on the fate of descendants of the thorium and uranium series, respectively (see Hevesy, [Gen76]).

**28.5. Rare-earth Elements (Lanthanide Series) and Other Elements in Fission.** The fission of uranium and plutonium results in the production of isotopes of elements ranging from number 30 (zinc) to number 63 (europium). Fission products therefore include species of almost half the rare-earth elements (numbers 57 to 71) or lanthanide series. Thus elements for which very little biological interest existed before the large-scale utilization of the fission process now have assumed considerable importance by virtue of the dangers of contamination by, and irradiation from, their radioactive species as produced in atomic-energy installations. A number of the elements occurring in large yields in fission have already been considered, *e.g.*, strontium, columbium, xenon, and iodine. Other elements occurring as fission products for which biological studies have been carried out include (*i.e.*, those not already considered previously) the rare earths lanthanum, cerium, praseodymium, and promethium and the elements selenium, yttrium, zirconium, ruthenium, tellurium, gallium, and barium. Studies with all these except selenium, yttrium, zirconium, and tellurium have been limited to one or two investigations concerned with fission-product problems.

No biological studies have as yet been carried out with radioactive species of the rare-earth elements not occurring in fission, nor of several fission elements of lighter weight (gallium, germanium, tellurium, rhodium, palladium, etc.).

Extensive work has been carried out by Hamilton and coworkers [Ba1,Ce1, 2, etc.] on the distribution of fission-product elements in rats, particularly in the liver and bone. Most of these are not taken up in significant amounts from the digestive tract after oral administration, exceptions being strontium, tellurium, iodine, barium, and cesium. Following parenteral injection, however, many are accumulated in the skeleton and eliminated very slowly: strontium, yttrium, zirconium, columbium, barium, lanthanum, cerium, praseodymium, and promethium, but not ruthenium, tellurium, iodine, or cesium. Initially lanthanum, cerium, praseodymium, and promethium accumulate in the liver, but are quite rapidly excreted from this organ. Of the fission elements concentrating in bone, those not taken into the body in appreciable amounts from the digestive tract are retained for long periods of time in the pulmonary tissue when administered intratracheally or by aerosols. Most of what is absorbed deposits in the bone. This has considerable significance in the public-health aspects of atomic-energy installations, especially as chronic radiation of bone by radioactive isotopes localized there has long been known for radium and recently demonstrated for strontium and plutonium [Sr15,Pu10] to cause osteosarcomas.

Studies on Se<sup>75</sup> metabolism have been restricted to two papers on the distribution of labeled selenate injected into the rat [Se1, 2]. It was found that selenate labeled with Se<sup>83</sup> (30-min half-life) is rapidly converted in the body into volatile organic compounds and eliminated in the breath. Studies on tellurium with the isotope Te<sup>121</sup> (125-day half-life) [Te1] and a mixture of the isotopes Te<sup>127</sup> (90-day half-life) and Te<sup>129</sup> (32-day half-life) [Te2] have shown, on the other hand, that at first the urine is the principal route of elimination and later is almost equal to the feces; tellurium is concentrated most highly in the blood, liver, and kidneys, apparently whether administered in the tellurous or telluric state.

Radioisotopes of yttrium and zirconium have been used in colloidal form in the study of blood mixing time and volume (Y2,Zr2). They also have found, or been suggested for, preliminary therapeutic application in the treatment of leukemias (see Chap. 29).

**28.6. New Rare-earth Elements (Actinide Series).** The new rare-earth elements or actinide series begins with actinium (element 89) and includes the heaviest known elements. So far eight have been discovered: actinium, thorium, protactinium, uranium, neptunium, plutonium, americium, and curium. Biological studies have been carried out for all except actinium (which, however, has species in the natural radioactive series, which thus

contribute to the radiotoxicity of the parent elements). No attempt has been made here to cover the extensive literature on the toxicology of thorium and uranium, nor on the application of thorium (as thorotrast) in radiography. Studies with the other five actinides have been recent and as yet rather limited. However, a great deal of work may be expected on plutonium, an element of which the isotope  $\text{Pu}^{239}$  is made artificially in large quantities in the uranium-pile reactor but which exists in nature only in minute traces.

Hamilton's comprehensive study of the metabolism of fission elements was done simultaneously with study of the six actinides—thorium, protactinium, neptunium, plutonium, americium, and curium [Pa1, etc.]. He has found that like lanthanum, germanium, praseodymium, and promethium they concentrate to a high degree in bone when administered parenterally, but are not absorbed in significant amounts from the digestive tract. Radioautographic study has demonstrated that bone deposition does not occur uniformly in the osseous tissue (as with strontium), but rather in the periosteum and endosteum, and in the region of the trabecular bone. Recent work has also shown that  $\text{Pu}^{239}$  (24,000-year half-life) in bone can produce malignant change [Pu10].

**28.7. Other Elements.** Elements not already covered in this chapter include two light elements, scandium and titanium, for which no tracer work has been done, and a number of the heavier elements above the lanthanide series,—namely, hafnium, tantalum, rhenium, osmium, iridium, platinum, thallium, polonium, astatine, and francium. Of the latter only polonium and astatine have been studied biologically.

Only two studies [Po1,2] on the distribution and biological effect of polonium have been carried out with the isotope  $\text{Po}^{210}$ , a member of the uranium series. The element concentrates in the reticuloendothelial system, especially of the spleen. The first autoradiographs were taken with the use of tissue slices containing polonium [Po1].

Astatine has been studied with the isotope  $\text{At}^{211}$  (7.5-hr half-life). In the one published paper dealing with this species [At1] it has been found to resemble its analogue iodine quite closely in being concentrated to a high degree in the thyroid.

## CHAPTER 29

### ISOTOPES IN THERAPY AND DIAGNOSIS

ELLSWORTH C. DOUGHERTY

**29.1. Introduction.** It has not seemed desirable here to survey exhaustively the use of radioactive isotopes in therapy and diagnosis. The material presented is merely designed to orient the reader in this general field. For more complete discussions see the review papers by Dougherty and Lawrence [Gen38,39] and the various papers cited in Table 49. Isotopes that have been used or suggested in therapy are given in Table 49.

Isotopes of radium (specifically  $\text{Ra}^{226}$ ) and radon (specifically  $\text{Rn}^{222}$ ) have been used for almost fifty years in the treatment of various dermatoses and malignancies. For the most part the effectiveness of these isotopes has depended upon the penetrating gamma rays arising from  $\text{Bi}^{214}$  ( $\text{RaB}$ ) and  $\text{Po}^{214}$  ( $\text{RaC}$ ), which are present in mixtures of  $\text{Ra}^{226}$  and  $\text{Rn}^{222}$  in equilibrium with decay products. Rarely, the penetrating beta particles from such mixtures may also be used in therapy. The alpha particles arising from various members of the radium-radon decay system are of negligible importance in this connection.  $\text{Ra}^{226}$  and  $\text{Rn}^{222}$  are today used only locally. In the first two decades of the century both found use systemically—injected, inhaled, or ingested in appropriate vehicles, but the results were not satisfactory. Although records are lacking, some of the individuals so treated must ultimately have suffered “radium” poisoning. Two other isotopes of radium,  $\text{Ra}^{223}$  ( $\text{ThX}$ ) and  $\text{Ra}^{223}$  ( $\text{MsTh}_1$ ), also found early systemic use.

Aside from isotopes of radium and radon and those occurring as their decay products the actually and potentially useful species are artificial ones—of the elements sodium, phosphorus, manganese, cobalt, zinc, strontium, yttrium, zirconium, columbium, iodine, gold, and most importantly hydrogen and carbon.

Although local application of artificial radioactive isotopes has been carried out and will undoubtedly play an important future role, in all likelihood replacing radium and radon as therapeutic agents, the particular importance of most useful species lies in their systemic application. Artificial isotopes that so far have shown definite value in therapy and diagnosis have been those that demonstrate a fair degree of selective localization, either as the result of their normal fate in metabolism or of their fate as special compounds or colloids. Thus phosphorus concentrates to a small degree

in rapidly growing tissue (particularly in the nucleoprotein) and in bone; iodine localizes in the thyroid gland (at least several hundred times the concentration in the rest of the body); and certain colloidal preparations can be made that localize in various parts of the reticuloendothelium.

So far, all techniques of radiation with isotopes, except in the case of the treatment of hyperthyroidism with radioiodine, have suffered from the limited selectivity of uptake by the diseased tissue over normal tissues. For example, in the treatment of diseases of the bone marrow and lymphatic system with radioactive phosphorus it is impossible to avoid to a greater or lesser extent the irradiation in various parts of the body of those normal cells that metabolize rapidly and others in the immediate vicinity of the latter. Actually in the treatment of polycythemia vera it would be desirable if irradiation could be delivered only to the nucleated red cells and in lymphosarcoma and leukemia only to the offending leukocytes or lymphosarcoma cells, and not to the normal cells, especially the platelet and red-cell-producing centers. Some progress may be anticipated in the management of these blood dyscrasias by the application of labeled colloids. Recent work [P228] shows that radiation may be limited largely to the liver and spleen by the use of anhydrous colloidal chromic phosphate labeled with  $P^{32}$ , which localizes selectively in the reticuloendothelial cells of the liver and spleen when introduced intravascularly in a highly dispersed form. Similar properties have been discovered [Zr3] for a number of other colloids—of yttrium, zirconium, and columbium—which may be labeled with radioisotopes of these respective elements. Some of the colloids can be concentrated to a high degree in the bone marrow, others in the liver and spleen.

However, for neoplastic diseases in general, localizing substances of adequate selectivity that can be labeled and used for radiotherapy have not been found.

The availability of radioactive isotopes for internal therapy has been critically evaluated recently by Hahn and Sheppard [Gen66a], who established the following criteria of therapeutic usefulness:

1. An isotope should be pure and free from contaminants.
2. The "half-life" should be less than 10 days and more than 2 days.
3. The physical and chemical properties must be known and the biological behavior understood.
4. Localization and selectivity in deposition are necessary.

Only 18 isotopes could be listed from the 450 known in 1944 (Seaborg's table). There are, however, other isotopes (*i.e.*,  $Co^{60}$ ), which may be useful by being removable after therapeutic application, just as is radium.

The above criteria must be met in part at least for isotopes to be used in diagnosis, but, because tracer rather than therapeutic amounts would in most cases be used, the potential limitations are scarcely comparable.



**29.2. Radiophosphorus.** The first artificial radioisotope to be used successfully in therapy was  $P^{32}$ . Although applied as early as 1936 it was not until 1939 that the first clinical reports [P249] of its application to the treatment of chronic leukemia were published.

The most important use of  $P^{32}$  has been in soluble form (in labeled isotonic disodium acid phosphate) given by mouth or vein in the treatment of blood dyscrasias. The determination and calculation of dosage, routes of administration, and supportive treatment of patients receiving this type of therapy are discussed in works cited in Table 49. Other uses of  $P^{32}$  have been in topical application [P265] for warts, angiomas, and other skin lesions, and in insoluble colloid anhydrous chromic phosphate for hepatosplenomegaly in leukemia and allied diseases.

The use of  $P^{32}$  as a possible diagnostic tool has been suggested by Low-Beer and associates [P266]. They have found in a few cases that, if small doses of labeled sodium phosphate are given to women with suspected breast carcinoma, the activity of the superficially located malignant lesion (except for slow-growing mucoid carcinomas) is 25 per cent or more above that of the surrounding tissue of the skin. Benign lesions in their series consistently showed less than 25 per cent greater activity.

The use of  $P^{32}$  has thus been shown to have considerable therapeutic and possible diagnostic value. Further research may well expand its clinical usefulness in the form of other inorganic or organic compounds.

**29.3. Radioiodine.** The therapeutic use of radioactive iodine was reported for the first time simultaneously in 1942 [I29,40] for the treatment of hyperthyroidism (*i.e.*, thyrotoxicosis, Graves' disease, Basedow's disease, etc.). Comprehensive reports on two series of patients with hyperthyroidism have appeared recently [I9,41].

For therapeutic purposes carrier-free radioiodine in the form of an aqueous sodium iodide solution is used, being administered by mouth. In the two most recent reports [I9,41], the iodine has consisted of a mixture of  $I^{130}$  and  $I^{131}$ , of which the former has been the clinically more important component. The determination and calculation of dosage are given in both papers.

In the last few years workers have also reported a few cases in which treatment of iodine-metabolizing thyroid malignancy has met with partial or considerable success when radioiodine has been used [I83,105,114].

As a diagnostic tool for the determination of the level of the thyroid function, radioiodine appears to offer good clinical possibilities. So far, however, most of the publications dealing with thyroid physiology as studied with radioiodine have been in the nature of fundamental investigations rather than of diagnostic developments.

In general the use of iodine for the treatment and diagnosis of certain thyroid diseases appears highly promising.

**29.4. Radiosodium.** The distribution of sodium primarily as an extracellular cation has permitted the application of ionic  $\text{Na}^{24}$  as a substitute for total irradiation in leukemia and allied diseases [Na58,81]. Patients so treated have shown symptomatic improvement and a fall in the blood count. It may prove as valuable as  $\text{P}^{32}$  and in some cases more advantageous in view of the greater ease with which dosage may be regulated because of the shorter half-life and more rapid excretion. Its use has yet to be thoroughly tested clinically.

The usefulness of radiosodium as a diagnostic tool is evident from a recent review [Na69] in which it is reported that in a number of circulatory diseases (arteriosclerosis, Raynaud's disease, etc.) and injuries (trench foot and immersion foot, frostbite, etc.) of the extremities  $\text{Na}^{24}$  has proved of considerable value in determining the status of the circulation. Since it can be measured by applying a counter to the surface of the skin and thus detecting its penetrating gamma rays, its distribution in the blood and extracellular-tissue fluids can be followed. With this technique of measuring tissue vascularity it has been possible to determine in a number of cases whether surgery is indicated and, if so, at what point the amputation should be performed. Similar possibilities exist with the use of radioactive inert gases. It has also been possible to measure blood-circulation time with  $\text{Na}^{24}$  [Na69].

**29.5. Radiocolloids.** A number of radioactive colloids have been used in preliminary therapeutic studies or suggested as therapeutically promising. These include, as already mentioned, anhydrous chromic phosphate [P228], which localizes almost entirely in the liver and spleen; colloidal manganese dioxide protected by gelatin [Mn7,10]; zinc suspended in a suitably prepared solution of pectin [Zn2-4]; colloidal sols of gold [Au2-4]; and colloids of several elements, radioisotopes of which occur as fission products [Gen38].

The fission colloids may, depending upon the method of preparation, concentrate upon intravenous administration almost exclusively in the liver and spleen or go in equal concentration into the bone marrow [Zr3]. Because of the relative abundance of yttrium, zirconium, and columbium in fission, there is a potentially rich source of their radioactive species for such investigations and for possible widespread clinical application. The criteria of Hahn and Sheppard [Gen66a], however, may rule out these fission elements as important radiotherapeutic agents, although shorter lived species such as  $\text{Y}^{90}$  (60-hr half-life) should prove very useful.

Investigations with radiomanganese as labeled colloidal manganese dioxide in an aqueous medium with gelatin as a protective colloid have recently been reported [Mn7,10,11]. These suggest the possibility that manganese dioxide may be localized upon intravenous injection in the lymphoid reticuloendothelium.

In addition to the intravascular administration of these colloids there are

also possible applications in the interstitial infiltration of malignant growths, for which chromic phosphate [P10], colloidal zinc [Zn2-4], and colloidal gold [Au2-4] have already been employed in preliminary work with promising results.

**29.6. Other Isotopes.** Although phosphorus, iodine, and sodium radioisotopes are the only artificial species for which therapeutic applications have been definitely developed, it seems likely that in a few years a number of different isotopes will have well-accepted clinical uses.

Radiostrontium has been suggested and in a few cases tried [Sr14] in the therapy of bone malignancy because the behavior of this element is analogous to that of calcium, which of course concentrates in bone. Although a comprehensive report is lacking, it seems unlikely that diffuse irradiation of bone and marrow for relatively localized malignant growths will prove of much value, particularly in view of the danger of marrow depression.

Recently Co<sup>60</sup> has been suggested as a substitute for radium. The fact that it can be used as the free metal, can be worked into the desired shape, and has a usefully long half-life makes this use an attractive possibility.

In addition to the elements mentioned there are important possibilities in the labeling of organic compounds with long-lived radiocarbon C<sup>14</sup> and with tritium H<sup>3</sup>. If artificial radioactivity is ever to prove a vital tool in the cure of malignancy, the only way for accomplishing this end would seem, from our present perspective, to be the discovery of organic substances that undergo highly selective localization in various types of cancer cells in contrast to those of normal tissue and that can be labeled with a suitable radioisotope. The use of C<sup>14</sup> must be approached with caution because of the possibility of its long-term deposition as carbonate in bone [C25] and the consequent danger of induced malignant degeneration. Tritium with the low penetrating power of its beta particles and its rapid turnover in the body is particularly promising because if it can be put in relatively stable positions in localizing compounds, it will expend most of its ionizing power within the concentrating cells. Potentially it is an ideal radiotherapeutic agent.

The therapeutic use of fissionable isotopes, such as U<sup>235</sup> and Pu<sup>239</sup>, and other species capable of releasing large amounts of ionizing radiation upon being bombarded with slow neutrons, such as Li<sup>6</sup> and B<sup>10</sup>, does not seem very promising because, even though it may prove possible to localize them at specific points in the body, the short mean path of thermal neutrons, the damaging effects of their infiltration, and the toxic effects of fissionable materials and their products all pose serious, if not insuperable, difficulties.

Finally there should be mentioned the possible therapeutic use of charged nuclear particles—protons, deuterons, and alpha particles—to high energies in the cyclotron and other heavy-particle accelerators. This would be an isotopic application of sorts inasmuch as the nuclei of single species of hydro-

TABLE 46. IMPORTANT STABLE AND RADIOACTIVE ISOTOPES THAT HAVE BEEN USED IN BIOLOGICAL AND MEDICAL INVESTIGATIONS: *Tracer Studies*

For extended discussions of tracer studies with radioactive isotopes, see books of Kamen [Gen80] and Hevesy [Gen76]. For discussion of use of stable isotopes, see book by Govaerts [Gen 62].

The elements marked with a dagger † (and their naturally occurring isotopes) are known or believed to be essential in mammalian nutrition.

An isotope is placed in parentheses if it occurs together with another isotope or isotopes (of the same element) when used for tracer work but is itself not significant for the tracer purposes to which the mixture may be applied. Two isotopes are connected by a brace if they occur together in tracer preparations and are of about equal importance.

## A. Stable isotopes

Z	Element	Isotope	Some important problems to which isotope has been applied
	Name		
1	Hydrogen†	H <sup>2</sup> (D)	Intermediary metabolism, body-water turnover and content
6	Carbon†	C <sup>13</sup>	Intermediary metabolism
7	Nitrogen†	N <sup>15</sup>	Intermediary metabolism
8	Oxygen†	O <sup>18</sup>	Photosynthesis; bacterial fractionation; sulfate metabolism
16	Sulfur†	S <sup>34</sup>	Intermediary metabolism

## B. Artificial radioactive isotopes

1	Hydrogen†	H <sup>3</sup> (T)	Body-water content; photosynthesis
4	Beryllium	Be <sup>7</sup>	Absorption, distribution, and excretion
6	Carbon†	C <sup>11</sup>	CO <sub>2</sub> metabolism in plants and animals; CO distribution and elimination
		C <sup>14</sup>	Photosynthesis; intermediary metabolism; mineral metabolism
7	Nitrogen†	N <sup>13</sup>	Nitrogen fixation by barley; respiratory gas exchange
9	Fluorine	F <sup>18</sup>	Bone and tooth physiology and chemistry
11	Sodium†	Na <sup>22</sup>	Retention in congestive heart disease; effect
		Na <sup>24</sup>	Electrolyte metabolism; adrenal physiology; therapy of leukemia
12	Magnesium†	Mg <sup>27</sup>	Photosynthesis
15	Phosphorus†	P <sup>32</sup>	Mineral metabolism; intermediary metabolism; diagnosis of malignancy; colloids localizing in the reticuloendothelial system
16	Sulfur†	S <sup>35</sup>	Intermediary metabolism; thiamine metabolism; mineral metabolism
17	Chlorine†	Cl <sup>38</sup>	Mineral metabolism
18	Argon	Ar <sup>41</sup>	Respiratory gas exchange; blood circulatory pattern
19	Potassium†	K <sup>42</sup>	Mineral metabolism; adrenal physiology
20	Calcium†	(Ca <sup>41</sup> ) Ca <sup>45</sup>	Mineral metabolism

TABLE 46. IMPORTANT STABLE AND RADIOACTIVE ISOTOPES THAT HAVE BEEN USED IN BIOLOGICAL AND MEDICAL INVESTIGATIONS: *Tracer Studies*—(Continued)

Element		Isotope	Some important problems to which isotope has been applied
Z	Name		
25	Manganese†	Mn <sup>52</sup> (Mn <sup>54</sup> ) (Mn <sup>55</sup> ) Mn <sup>56</sup> Mn <sup>58</sup>	Distribution of colloidal MnO <sub>2</sub> in reticuloendothelial system Mineral metabolism
26	Iron†	Fe <sup>55</sup> (Fe <sup>56</sup> ) Fe <sup>57</sup> Fe <sup>59</sup>	Mineral metabolism Mineral metabolism; blood physiology; blood preservation Mineral metabolism; blood physiology; blood preservation Mineral metabolism; blood physiology
27	Cobalt†	Co <sup>56</sup> (Co <sup>57</sup> ) Co <sup>58</sup> Co <sup>60</sup>	Mineral metabolism Mineral metabolism
29	Copper†	Cu <sup>64</sup>	Mineral metabolism
30	Zinc†	Zn <sup>65</sup> Zn <sup>66</sup>	Distribution of injected colloidal zinc Mineral metabolism
33	Arsenic	As <sup>74</sup>	Absorption, distribution, and excretion; experimental filariasis
34	Selenium	Se <sup>75</sup>	Absorption, distribution, and excretion
35	Bromine	Br <sup>82</sup>	Electrolyte exchange; thyroid physiology; distribution of brominated dyes
36	Krypton	Kr <sup>79,81</sup>	Respiratory gas exchange; blood circulatory pattern
37	Rubidium	Rb <sup>86</sup>	Electrolyte exchange
38	Strontium	Sr <sup>85</sup> Sr <sup>86</sup> Sr <sup>87</sup>	Absorption, distribution, and excretion§ Bone physiology Absorption, distribution, and excretion
39	Yttrium	Y <sup>91</sup>	Absorption, distribution, and excretion; fate of colloids in reticuloendothelial system
40	Zirconium	Zr <sup>90</sup> Zr <sup>91</sup>	Absorption, distribution, and excretion§ Absorption, distribution, and excretion; fate of colloids in reticuloendothelial system
41	Columbium	Cb <sup>95</sup>	Absorption, distribution, and excretion; fate of colloids in reticuloendothelial system
42	Molybdenum†	Mo <sup>98</sup> (Mo <sup>99</sup> ) Mo <sup>100</sup>	Absorption by plants Absorption, distribution, and excretion; absorption in plants
4	Ruthenium	Ru <sup>100</sup> Ru <sup>101</sup>	Absorption, distribution, and excretion
51	Antimony	Sb <sup>124</sup>	Absorption, distribution, and excretion; experimental filariasis
52	Tellurium	Te <sup>122</sup> Te <sup>123</sup> (Te <sup>126</sup> )	Absorption, distribution, and excretion Absorption, distribution, and excretion

TABLE 46. IMPORTANT STABLE AND RADIOACTIVE ISOTOPES THAT HAVE BEEN USED IN BIOLOGICAL AND MEDICAL INVESTIGATIONS: *Tracer Studies*—(Continued)

Element Z Name	Isotope	Some important problems to which isotope has been applied
53 Iodine†	I <sup>128</sup> I <sup>130</sup> (I <sup>130</sup> ) I <sup>131</sup>	Iodine metabolism and thyroid physiology  Iodine metabolism and thyroid physiology; absorption of insulin
54 Xenon	Xe <sup>127</sup>  Xe <sup>133</sup>	Respiratory gas exchange; blood circulatory pattern  Absorption and distribution
55 Cesium	Cs <sup>134</sup> Cs <sup>137</sup>	Absorption, distribution, and excretion§ Absorption, distribution, and excretion
56 Barium	Ba <sup>133</sup> Ba <sup>140</sup>	Absorption, distribution, and excretion§ Absorption, distribution, and excretion
57 Lanthanum	La <sup>140</sup>	Absorption, distribution, and excretion
58 Cerium	Ce <sup>141</sup> Ce <sup>144</sup>	Absorption, distribution, and excretion  Absorption, distribution, and excretion
59 Praseodymium	Pr <sup>143</sup>	Absorption, distribution, and excretion
61 Promethium	Pm <sup>147</sup>	Absorption, distribution, and excretion
79 Gold	Au <sup>198</sup>	Absorption, distribution, and excretion; chrysotherapy in arthritis
80 Mercury	Hg <sup>197</sup>	Mercury vapor as industrial health hazard
85 Astatine	At <sup>211</sup>	Thyroid physiology
91 Protoactinium	Pa <sup>233</sup>	Absorption, distribution, and excretion
93 Neptunium	Np <sup>239</sup>	Absorption, distribution, and excretion
94 Plutonium	Pu <sup>239</sup>	Absorption, distribution, and excretion
95 Americium	Am <sup>241</sup>	Absorption, distribution, and excretion
96 Curium	Cm <sup>242</sup>	Absorption, distribution, and excretion

## C. Natural radioactive isotopes

82 Lead	Pb <sup>210</sup> (RaD) Pb <sup>212</sup> (ThB)	Absorption, distribution, and excretion Absorption, distribution, and excretion
83 Bismuth	Bi <sup>210</sup> (RaE) Bi <sup>214</sup> (RaC)	Absorption, distribution, and excretion Velocity of blood flow
84 Polonium	Po <sup>210</sup>	Distribution and excretion
86 Radon	Rn <sup>220</sup> (Th) Rn <sup>222</sup>	Elimination in breath Numerous studies on uptake and elimination in breath, distribution, etc.
88 Radium	Ra <sup>224</sup> (ThX) Ra <sup>226</sup> Ra <sup>228</sup> (MsTh <sub>1</sub> )	Distribution Numerous studies on distribution Distribution
90 Thorium	Th <sup>232</sup> (RdTh) Th <sup>232</sup> (natural element)	Absorption, distribution, and excretion Numerous studies on distribution  Absorption, distribution, and excretion
92 Uranium	Th <sup>234</sup> (UX <sub>1</sub> ) (U <sup>235</sup> ) U <sup>238</sup> (natural element)	Numerous studies on distribution    Numerous studies on distribution

§ Personal communication from Dr. J. G. Hamilton.

TABLE 47. IMPORTANT STABLE AND RADIOACTIVE ISOTOPES THAT HAVE BEEN USED IN BIOLOGICAL AND MEDICAL INVESTIGATIONS: *Nontracer Studies*

The naturally occurring isotopes of elements having but one stable species are not listed here, except for Be<sup>9</sup> which has been applied to an irradiation problem. Although it is obvious that any use of the natural element in such cases is a use of a single isotope, neither tracer nor radiation techniques are involved (with the exception noted).

(For more details of therapeutic applications, see Table 49.)

Element Z Name	Isotope	Some important problems to which isotope has been applied
A. Stable isotopes		
1 Hydrogen	H <sup>1</sup>	High-energy proton irradiation (suggested)
	H <sup>2</sup>	High-energy deuteron irradiation
2 Helium	He <sup>4</sup>	Alpha-particle irradiation
3 Lithium	Li <sup>6</sup>	Slow neutron irradiation of Li-infiltrated tumors
4 Beryllium	Be <sup>9</sup>	Fast neutron irradiation and therapy
	(natural element)	
5 Boron	B <sup>10</sup>	Slow neutron irradiation of B-infiltrated tumors
B. Artificial radioactive isotopes		
1 Hydrogen	H <sup>3</sup>	Therapy (suggested)
6 Carbon	C <sup>14</sup>	Therapy (suggested)
11 Sodium	Na <sup>24</sup>	Therapy
15 Phosphorus	P <sup>32</sup>	Therapy; as beta-ray source in irradiation experiments
25 Manganese	Mn <sup>52</sup>	Therapy (suggested)
	(Mn <sup>54</sup> )	
27 Cobalt	Co <sup>60</sup>	As radium substitute in therapy (suggested)
30 Zinc	Zn <sup>63</sup>	Preliminary therapy
38 Strontium	Sr <sup>89</sup>	Therapy
39 Yttrium	Y	Therapy
40 Zirconium	Zr	Therapy (suggested)
41 Columbium	Cb	Therapy (suggested)
42 Iodine	I <sup>130</sup>	Therapy
	(I <sup>131</sup> )	
	(I <sup>130</sup> )	Therapy
	I <sup>131</sup>	
79 Gold	Au <sup>198</sup>	Preliminary therapy
C. Natural radioactive isotopes		
81 Thallium	Th <sup>230</sup> (ThC'')	(See Ra <sup>226</sup> )
	Th <sup>230</sup> (RaC'')	(See Ra <sup>226</sup> )
82 Lead	Pb <sup>212</sup> (ThB)	(See Ra <sup>226</sup> )
	Pb <sup>214</sup> (RaB)	(See Ra <sup>226</sup> )
83 Bismuth	Bi <sup>212</sup> (ThC)	(See Ra <sup>226</sup> )
	Bi <sup>214</sup> (RaC)	(See Ra <sup>226</sup> )

TABLE 47. IMPORTANT STABLE AND RADIOACTIVE ISOTOPES THAT HAVE BEEN USED IN BIOLOGICAL AND MEDICAL INVESTIGATIONS: *Nontracer Studies—(Continued)*

Element	Isotope	Some important problems to which isotope has been applied
Z Name		
84 Polonium	Po <sup>212</sup> (ThC')	(See Ra <sup>224</sup> )
	Po <sup>214</sup> (RaC')	(See Ra <sup>226</sup> )
	Po <sup>216</sup> (ThA)	(See Ra <sup>224</sup> )
	Po <sup>218</sup> (RaA)	(See Ra <sup>226</sup> )
86 Radon	Rn <sup>220</sup> (Tn)	(See Ra <sup>224</sup> )
	Rn <sup>222</sup>	Therapy
88 Radium	Ra <sup>224</sup> (ThX)	Therapy (abandoned)
	Ra <sup>226</sup>	Therapy
	Ra <sup>228</sup> (MsTh <sub>1</sub> )	Therapy (abandoned)
89 Actinium	Ac <sup>228</sup> (MsTh <sub>2</sub> )	(See Ra <sup>228</sup> )
92 Uranium	U <sup>235</sup>	Slow neutron irradiation; fission of U colloids in reticuloendothelial system

gen and helium are being employed. None of these has been used clinically as yet.

By comparison with their therapeutic limitations, the use of radioactive and stable isotopes in diagnosis is far more promising. There is probably no clinical condition for which a useful application of an isotope might not be worked out. It will not be surprising if in 25 years there are scores of standard diagnostic procedures employing isotopes of most of the elements of the periodic system.



TABLE 48. POTENTIALLY VALUABLE TRACER ISOTOPES, ESPECIALLY THOSE OF ELEMENTS FOR WHICH BIOLOGICAL STUDIES HAVE NOT YET BEEN CARRIED OUT

The elements marked with an asterisk\* appear in Table 46.

In connection with the rarer stable species listed here it should be understood that the one to several more abundant species, when prepared in pure form or enriched, may in some cases find application as tracers, either independently or, in the case of multiple labeling experiments, in conjunction with the rarer species.

Element	Isotope	Half-life (or per cent abundance)	Element	Isotope	Half-life (or per cent abundance)†
Z Name			Z Name		
2 Helium	He <sup>3</sup>	Stable ( $1.3 \times 10^{-5}$ )	21 Scandium	Sc <sup>44</sup>	2.44 d
3 Lithium	Li <sup>6</sup>	Stable (7.39)		Sc <sup>46</sup>	85 d
4 Beryllium*	Be <sup>10</sup>	( $2.5 \times 10^6$ y)	22 Titanium	Sc <sup>48</sup>	44 h
5 Boron	B <sup>10</sup>	Stable (18.83)		Ti <sup>44</sup>	3.08 h
8 Oxygen*	O <sup>15</sup>	126 s	23 Vanadium	Ti <sup>46</sup>	72 d
	O <sup>17</sup>	Stable (0.039)		V <sup>48</sup>	16 d
10 Neon	Ne <sup>21</sup>	Stable (0.27)	24 Chromium	V <sup>(48)</sup>	600 d
	Ne <sup>22</sup>	Stable (9.73)		Cr <sup>48</sup>	41.9 m
12 Magnesium*	Mg <sup>25</sup>	Stable (10.11)	26 Iron*	Cr <sup>51</sup>	26.5 d
	Mg <sup>26</sup>	Stable (11.29)		Fe <sup>52</sup>	7.8 h
13 Aluminum	Al <sup>26</sup>	2.30 m		Fe <sup>54</sup>	Stable (5.81)
	Al <sup>28</sup>	6.7 m		Fe <sup>57</sup>	Stable (2.20)
14 Silicon	Si <sup>29</sup>	Stable (4.67)	27 Cobalt*	Fe <sup>58</sup>	Stable (0.33)
	Si <sup>30</sup>	Stable (3.05)	28 Nickel	Co <sup>56</sup>	18.2 h
16 Sulfur*	Si <sup>31</sup>	170 m		Ni <sup>57</sup>	36 h
	S <sup>32</sup>	Stable (0.74)		Ni <sup>(58)</sup>	12 y
	S <sup>36</sup>	Stable (0.016)	29 Copper*	Ni <sup>60</sup>	56 h
17 Chlorine*	Cl <sup>36</sup>	$2 \times 10^6$ y		Cu <sup>61</sup>	3.4 h
	Cl <sup>37</sup>	Stable (24.6)		Cu <sup>65</sup>	Stable (30.91)
18 Argon*	A <sup>37</sup>	34.1 d	30 Zinc*	Cu <sup>(67)</sup>	56 h
19 Potassium*	K <sup>40</sup>	$1.42 \times 10^9$ y (0.012)		Zn <sup>62</sup>	9.5 h
	K <sup>41</sup>	Stable (6.7)	31 Gallium	Zn <sup>66</sup>	13.8 h
20 Calcium*	K <sup>(43)</sup>	22.4 h		Zn <sup>72</sup>	49 h
	Ca <sup>42</sup>	Stable (0.64)	32 Germanium	Ga <sup>66</sup>	9.4 h
	Ca <sup>43</sup>	Stable (0.15)		Ga <sup>67</sup>	78.3 h
	Ca <sup>44</sup>	Stable (2.06)		Ga <sup>72</sup>	14.3 h
	Ca <sup>45</sup>	Stable (0.0033)	33 Arsenic*	Ga <sup>(74)</sup>	9 d
	Ca <sup>46</sup>	Stable (0.19)		Ge <sup>(68)</sup>	250 d
	Ca <sup>48</sup>	2.5 h		Ge <sup>71</sup>	11 d
				Ge <sup>71</sup>	39.7 h
				Ge <sup>77</sup>	12 h
			34 Selenium*	As <sup>(72)</sup>	26 h
				As <sup>(74)</sup>	90 d
				As <sup>76</sup>	26.8 h
				As <sup>(77)</sup>	40 h
			35 Bromine*	Se <sup>(72)</sup>	9.5 d
				Se <sup>(73)</sup>	6.7 h
				Se <sup>(78)</sup>	127 d
			36 Krypton*	Br <sup>(77)</sup>	48 h
				Br <sup>80m</sup>	4.4 h
			37 Rubidium*	Kr <sup>85</sup>	4.5 h
				Kr <sup>(85)</sup>	9.4 y
				Rb <sup>82</sup>	6 h
				Rb <sup>(84)</sup>	~ 40 d
			38 Strontium*	Sr <sup>(88)</sup>	2.7 h

TABLE 48. POTENTIALLY VALUABLE TRACER ISOTOPES, ESPECIALLY THOSE OF ELEMENTS FOR WHICH BIOLOGICAL STUDIES HAVE NOT YET BEEN CARRIED OUT—(Continued)

Element			Element		
Z	Name	Isotope	Z	Name	Isotope
		(or per cent abundance)			(or per cent abundance)†
39	Yttrium*	<sup>87</sup> Y 80 h	54	Xenon*	<sup>135</sup> Xe 9.2 h
		<sup>90</sup> Y 62 h	55	Cesium*	<sup>131</sup> Cs (131) 10.2 d
40	Zirconium*	<sup>90</sup> Zr (97) 17.0 h			<sup>132</sup> Cs (132) 7.1 d
41	Columbium*	<sup>91</sup> Cb (90) 18 h			<sup>134</sup> Cs (134) 2.3 y
		<sup>91</sup> Cb (91) 60 d			<sup>135</sup> Cs (135) 13 d
		<sup>92</sup> Cb 10.1 d	56	Barium*	<sup>131</sup> Ba (131) 12.0 d
		<sup>94</sup> Cb (94) 3 d	57	Lanthanum*	<sup>135</sup> La (135) 19.5 h
43	Technetium	<sup>96</sup> Tc (96) 52 d			<sup>141</sup> La (141) 3.7 h
		<sup>96</sup> Tc 20.0 h	58	Cerium*	<sup>135</sup> Ce (135) ~ 16 h
		<sup>98</sup> Tc (98) 4.30 d			<sup>137</sup> Ce (137) 36 h
		<sup>97m</sup> Tc 90 d			<sup>139</sup> Ce (139) 140 d
		<sup>99</sup> Tc $9.4 \times 10^5$ y			<sup>143</sup> Ce 33 h
44	Ruthenium*	<sup>97</sup> Ru 2.8 d	59	Praseodymium*	<sup>142</sup> Pr (142) 19.3 h
		<sup>100</sup> Ru (100) 4.5 h	60	Neodymium	<sup>147</sup> Nd (147) 12.1 d
45	Rhodium	<sup>100</sup> Rh (100) 19.4 h			<sup>149</sup> Nd (149) 1.8 h
		<sup>101</sup> Rh (101) 4.3 d	61	Promethium*	<sup>143</sup> Pm (143) ~ 200 d
		<sup>102</sup> Rh 210 d			<sup>147</sup> Pm ~ 4 y
		<sup>105</sup> Rh 36.5 h			<sup>148</sup> Pm 5.3 d
46	Palladium	<sup>106</sup> Pd (106) 4.0 d			<sup>149</sup> Pm 55 h
		<sup>101</sup> Pd 9 h	62	Samarium	<sup>151</sup> Sm (151) ~ 20 y
		<sup>102</sup> Pd 17 d			<sup>153</sup> Sm 47 h
		<sup>109</sup> Pd 13 h	63	Europium	<sup>152</sup> Eu (152) 9.2 h
		<sup>112</sup> Pd 21 h			<sup>154</sup> Eu 20 y
47	Silver	<sup>106</sup> Ag 8.2 d			<sup>155</sup> Eu 2-3 y
		<sup>110</sup> Ag 225 d			<sup>156</sup> Eu 15.4 d
		<sup>111</sup> Ag 7.5 d			<sup>157</sup> Eu (157) 15.4 h
		<sup>112</sup> Ag 3.2 h	64	Gadolinium	<sup>152</sup> Gd (152) 155-70 d
48	Cadmium	<sup>107</sup> Cd 6.7 h			<sup>160</sup> Tb 3.9 h
		<sup>109</sup> Cd 330 d	65	Terbium	<sup>160</sup> Tb 73.5 d
		<sup>115</sup> Cd 2.33 d			<sup>165</sup> Dy 145 m
		<sup>116</sup> Cd 43 d	66	Dysprosium	<sup>162</sup> Ho (162) 65 d
49	Indium	<sup>111</sup> In (111) 2.7 d			<sup>166</sup> Ho 27.0 h
		<sup>114</sup> In 48 d	68	Erbium	<sup>169,171</sup> Er (169,171) 9.4 d
		<sup>115m</sup> In 4.42 h			<sup>171</sup> Er (171) 7.5 h
50	Tin	<sup>113</sup> Sn 105 d	69	Thulium	<sup>166</sup> Tm (166) 7.7 h
		<sup>119m</sup> Sn (119m) 13 d			<sup>167</sup> Tm (167) 9.6 d
		<sup>121</sup> Sn 28 h			<sup>168</sup> Tm (168) ~ 150 d
		<sup>121,123</sup> Sn (121,123) 130 d			<sup>170</sup> Tm 127 d
51	Antimony*	<sup>120</sup> Sb (120) 6.0 d			<sup>171</sup> Tm (171) 500 d
		<sup>122</sup> Sb 2.8 d	70	Ytterbium	<sup>169</sup> Yb (169) 33 d
		<sup>125</sup> Sb (125) 2.7 y			<sup>175</sup> Yb 99 h
		<sup>127</sup> Sb 93 h	71	Lutecium	<sup>170</sup> Lu (170) 2.15 d
		<sup>128</sup> Sb 4.2 h			<sup>173</sup> Lu (173) 9 d
52	Tellurium*	<sup>118</sup> Te (118) 6.0 d			<sup>172</sup> Lu (172) > 100 d
		<sup>119</sup> Te (119) 4.5 d			<sup>176m</sup> Lu (176m) 3.67 h
		<sup>132m</sup> Te 30 h			<sup>177</sup> Lu 6.8 d
		<sup>133</sup> Te (133) 77 h	72	Hafnium	<sup>181</sup> Hf (181) 46 d
53	Iodine*	<sup>124</sup> I 4.0 d	73	Tantalum	<sup>176</sup> Ta (176) 8.0 h
		<sup>125</sup> I (125) 56 d			<sup>177</sup> Ta (177) 2.66 d
		<sup>126</sup> I 13.0 d			<sup>178</sup> Ta (178) 16 d
		<sup>132</sup> I (132) 2.4 h			<sup>180</sup> Ta 8.2 h
		<sup>133</sup> I (133) 22 h			<sup>182</sup> Ta 117 d
		<sup>135</sup> I 6.7 h			

TABLE 48. POTENTIALLY VALUABLE TRACER ISOTOPES, ESPECIALLY THOSE OF ELEMENTS FOR WHICH BIOLOGICAL STUDIES HAVE NOT YET BEEN CARRIED OUT—(Continued)

Element			Half-life (or per cent abundance)	Element			Half-life (or per cent abundance)
Z	Name	Isotope		Z	Name	Isotope	
74	Tungsten	W <sup>(181)</sup>	140 d	82	Lead*	Pb <sup>(203)</sup>	52 h
		W <sup>(185)</sup>	73.2 d			Pb <sup>(209)</sup>	3.32 h
		W <sup>(187)</sup>	24.1 h	83	Bismuth*	Bi <sup>(204)</sup>	12 h
75	Rhenium	Re <sup>(187)</sup>	64.0 h			Bi <sup>(206)</sup>	6.4 d
		Re <sup>(184)</sup>	50 d	84	Polonium*	Po <sup>(206)</sup>	9 d
		Re <sup>(186)</sup>	92.8 h			Po <sup>(207)</sup>	5.7 h
		Re <sup>(188)</sup>	18.9 h			Po <sup>(208)</sup>	3 y
76	Osmium	Os <sup>(186)</sup>	97 d	85	Astatine*	At <sup>(210)</sup>	8.3 h
		Os <sup>(191)</sup>	32 h	87	Francium	Fa <sup>(223)</sup>	21 m
		Os <sup>(194)</sup>	15.0 d	88	Radium	Ra <sup>(223)</sup>	11.2 d
77	Iridium	Ir <sup>(190)</sup>	10.7 d			Ra <sup>(226)</sup>	14.8 d
		Ir <sup>(192)</sup>	70 d	89	Actinium	Ac <sup>(224)</sup>	2.5 h
		Ir <sup>(194)</sup>	19.0 h			Ac <sup>(226)</sup>	10.0 d
78	Platinum	Pt <sup>(190)</sup>	4.33 d			Ac <sup>(227)</sup>	21.7 y
		Pt <sup>(197)</sup>	18 h			Ac <sup>(228)</sup>	6.13 h
		Pt <sup>(197)</sup>	3.3 d	90	Thorium*	Th <sup>(230)</sup>	30.9 m
79	Gold*	Au <sup>(198)</sup>	15.8 h			Th <sup>(230)</sup>	7,000 y
		Au <sup>(194)</sup>	39.5 h			Th <sup>(231)</sup>	25.5 h
		Au <sup>(196)</sup>	195 d			91	Protactinium*
		Au <sup>(196)</sup>	13 h	Pa <sup>(229)</sup>	1.5 d		
		Au <sup>(196)</sup>	5.55 d	Pa <sup>(230)</sup>	17.7 d		
		Au <sup>(199)</sup>	3.3 d	Pa <sup>(231)</sup>	3.43 X 10 <sup>4</sup> y		
80	Mercury*	Hg <sup>(201, 203)</sup>	51.5 d	Pa <sup>(232)</sup>	1.32 d		
81	Thallium	Tl <sup>(200)</sup>	27 h	92	Uranium*	U <sup>(230)</sup>	20.8 d
		Tl <sup>(202)</sup>	11.8 d			U <sup>(231)</sup>	4.2 d
		Tl <sup>(204)</sup>	2.7 y			U <sup>(232)</sup>	70 y
						U <sup>(237)</sup>	6.8 d

TABLE 49. ISOTOPES USED OR SUGGESTED IN THERAPY  
This list does not include the early use of natural radioactive isotopes.

Element Z Name	Iso- tope	Vehicle (and route of administration)	Disease treated	Clinical value	References
1 Hydrogen	H <sup>1</sup>	As high-energy proton beam	None (suggested for deep-seated cancer)		
6 Carbon	H <sup>3</sup>	Localizing organic preparations	None (theoretically for cancer)	Palliative and curative	Gen38,39
	C <sup>14</sup>	Localizing organic preparations	None (theoretically for cancer)		
11 Sodium	Na <sup>24</sup>	Ionic Na <sup>+</sup> (in NaCl solution by mouth or vein)	Leukemia	Palliative	Na58,81
15 Phosphorus	P <sup>32</sup>	Soluble labeled phosphate (by mouth or vein)	Leukemia and allied diseases; polycythemia vera	Palliative	P87,114,115, 172,173,221, 257,267,317, 378
		Labeled CrPO <sub>4</sub> (by vein)	Hepatosplenomegaly in leukemia	Palliative	Gen38,39
		Labeled phosphate solution (topical)	Warts, angiomas, basal-cell carcinomas	Curative	P265
25 Manganese	Mn <sup>52</sup>	MnO <sub>2</sub> colloid protected by gelatin (by vein)	Lymphoblastomas	Not established	Mn7,10
27 Cobalt	Co <sup>60</sup>	Cobalt wire (inserted locally)	None (suggested as radium substitute)		
30 Zinc	Zn <sup>65</sup>	Colloid protected by pecten (interstitially)	Various malignancies	Not established	Zn2-4
38 Strontium	Sr <sup>65</sup>	Strontium lactate (by vein)	Bone malignancy	Not established	Sr14,20
39 Yttrium	Y <sup>90</sup>	Yttrium lactate colloid (by vein)	Leukemia, polycythemia vera	Palliative	Gen38,39
	Y <sup>91</sup>	As with Y <sup>90</sup>	Leukemia, polycythemia vera	Palliative	(Personal communication of J. W. Gofman)
40 Zirconium	Zr <sup>95</sup>	Zirconium lactate colloid (by vein)	None (suggested for leukemia, etc.)		
41 Columbium	Cb <sup>95</sup>	Columbium lactate colloid (by vein)	None (suggested for leukemia, etc.)		

TABLE 49. ISOTOPES USED OR SUGGESTED IN THERAPY—(Continued)

Element Z Name	Iso- tope	Vehicle (and route of administration)	Disease treated	Clinical value	References
53 Iodine	$I^{130}$ ( $I^{131}$ )	Tracer-free iodide (by mouth or vein)	Hyperthyroidism	Curative	I9,29,40-42,78
	( $I^{130}$ ) $I^{131}$	Tracer-free iodide (by mouth or vein)	Hyperthyroidism and metastatic thyroid malignancy	Palliative, in some cases pos- sibly cura- tive	I9,29,40-42, 78,114,115
79 Gold	$Au^{198}$	Gold sols (by vein and interstiti- ally)	Lymphoblastom- as and local malignancy	Not estab- lished	Au2,4
86 Radon (and decay prod- ucts)	$Rn^{222}$	Radon seeds, ointments, etc. (applied locally)	Various derma- toses and ma- lignancies	Palliative or cura- tive	
88 Radium	$Ra^{226}$	Radium needles, etc. (inserted locally)	Various derma- toses and malig- nacies	Palliative or curative	

## CHAPTER 30

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### 30.23. Hydrogen

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